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CONTENTS

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CONTENTS

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*Extranuclear Effects on Angular Correlations of Nuclear Radiations**

By ROLF M. STEFFEN

Department of Physics, Purdue University, Lafayette, Indiana, U.S.A.

CONTENTS

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Part I. Theory

§ 1. Introduction

The rapid improvement of the experimental techniques of nuclear spectroscopy in recent years, together with the development of the nuclear shell model[1-4] and the unified (collective motion) nuclear model[5,6], has provided us with important knowledge of the properties of low-lying nuclear levels. Information about the dynamic characteristics, such as beta and gamma transitions between these levels, as well as knowledge of the static properties of the ground and of the first few excited states, such as angular momenta and parities[7,8], magnetic moments[9-12] and electric quadrupole moments[13-16], are of considerable importance in the refinement of these nuclear models.

One of the most valuable tools of nuclear spectroscopy for the determination of angular momenta and parities of excited states is the study of the angular correlation[17-22] and the polarization direction correlation[23] of successive nuclear radiations emitted from these states. In some cases the angular correlation is altered by electric and magnetic fields acting on the electric and magnetic moments of the nucleus. These fields may be externally applied fields or may be due to atomic, molecular, or crystal structure. A measurement of the magnetic moment and the electric quadrupole moment of excited states then becomes possible. Moreover, the dependence of the angular correlation on the chemical and physical state of the radioactive source can provide information about the electric and magnetic fields prevailing at the decaying nucleus, and thus could give information on atomic and molecular structure and the structure of liquids and solids just as do nuclear magnetic resonance absorption and nuclear induction[24-27].

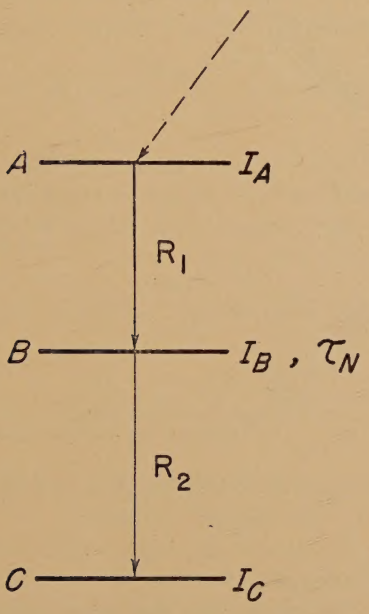
§ 2. The Unperturbed Angular Correlation of Nuclear Radiation

The following theoretical discussion of the general aspects of the angular correlation phenomenon without extranuclear perturbation is very brief and is intended only to introduce the notations used later. For a more detailed and comprehensive survey of the general theory of the angular correlation process the reader is referred to recent articles on this subject[28,40].

Consider a simple cascade of two nuclear transitions involving three levels A, B, C with angular momentum quantum numbers I_A, I_B, I_C (fig. 1). The intensity distribution of one radiation alone is of course isotropic in space due to the random orientation of the nuclear spin of the emitting nuclear levels. However, anisotropic radiation can arise when certain orientations of the nuclear angular momentum $\mathbf{I}\hbar$ are preferred with respect to some axis or, in other words, when there is unequal population of the magnetic sublevels of the emitting nuclear state, for some axis of quantization.

Such an unequal population of the m-states is introduced by the observation of the preceding radiation along a fixed direction $\mathbf{k}_1$, giving an ensemble of nuclei with unequal population of magnetic sublevels, when the fixed direction is taken as axis of quantization. The anisotropic intensity distribution of the second radiation with respect to the direction of propagation of the first one is known as the *angular correlation* of the two nuclear radiations. A necessary condition for such an anisotropic angular correlation to occur is that some unequal m-population in the intermediate state is preserved until the second radiation is emitted. In case no change of the magnetic sublevel population takes place during the intermediate state's lifetime, the correlation is termed ' undisturbed angular correlation '. This requires, in general, a rather short lifetime τ_N ($<10^{-10}$ sec) of the intermediate nuclear state, as we shall see later.

Fig. 1

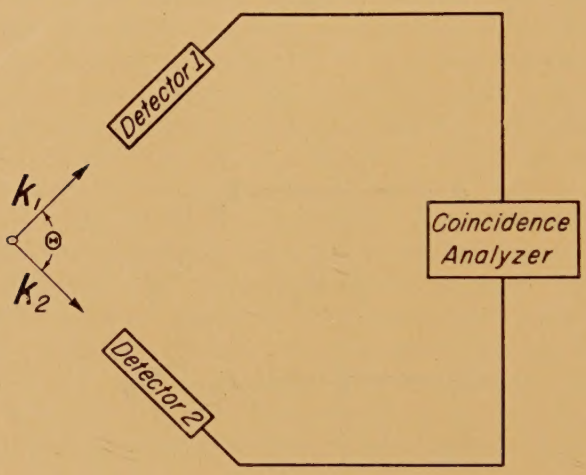


Two nuclear radiations emitted in cascade.

The experimental arrangement for the study of such an angular correlation is, in principle at least, quite simple. Two detectors D_1 and D_2 (fig. 2) record the two cascade radiations, emitted at an angle Θ with respect to each other, and a coincidence analyser selects only those nuclear quanta which are emitted simultaneously, or more precisely, which strike the detectors within a very short time interval τ_0 (10^{-9} sec to 10^{-6} sec), the coincidence resolving time. In this way only genetically related radiations, that is radiations emitted by the same nucleus, have

a good chance to be recorded. The dependence of the coincidence rate on the angle Θ (after some corrections for accidental coincidences, for the finite size of the detectors and the source, and for scattered radiation) is the measured angular correlation of the two nuclear radiations. The angular correlation of a particular nuclear cascade depends upon the angular momentum quantum numbers I_A, I_B, I_C of the three nuclear levels involved[29,30] and upon the types of radiations emitted, e.g. alpha rays[31,32], beta rays[33,34], gamma radiation[35], conversion electrons[36,37]. The fact that the correlation is also a sensitive function of finer details of nuclear transitions, such as the degree of forbiddenness and the type of interaction of beta transitions[33,34] and the multipole character of gamma transitions[29,38,39], makes it one of the most useful tools of nuclear spectroscopy.

Fig. 2



Schematic diagram of the experimental arrangement used for the measurement of an angular correlation.

For an unperturbed angular correlation the corresponding correlation function $W_0(\Theta)$, which expresses the relative probability* that the nuclear radiation R_2 is emitted at an angle Θ with respect to the propagation direction $\mathbf{k}_1$ of the first radiation R_1, is most conveniently expressed as a finite series of even Legendre polynomials :

$$W_0(\Theta)=1+\sum_{k=1}^{k_{\max}} A_{2k}P_{2k}(\cos\Theta), \quad \ldots\ldots \quad (1)$$

where $k_{\max}$ is the smallest integer of the set $(L_1)_{\max}$, $(L_2)_{\max}$ and I_B. $(L_1)_{\max}\hbar$ and $(L_2)_{\max}\hbar$ are the maximum values of the angular momentum

* Since $W(\Theta)$ is a relative probability, Θ-independent factors are irrelevant. Use is made of this fact to express $W(\Theta)$ in the most convenient form.

carried away by the nuclear radiation in the first and second transition, respectively. For the majority of nuclear cascades $k_{\max} \leqslant 2$, since angular momentum changes larger than $\Delta I = 2$ in the second transition would result in lifetimes τ_N of the intermediate state, which are too long to make an angular correlation experiment feasible.

The coefficients A_{2k} depend upon the aforementioned dynamic and static properties of the nuclear levels. They can be separated into two factors :

$$A_{2k} = A_{2k}^{(\mathrm{I})}(AB) \cdot A_{2k}^{(\mathrm{II})}(BC). \quad . \quad . \quad . \quad . \quad . \quad (2)$$

Each of these factors depends upon only one of the transitions $A \rightarrow B$ and $B \rightarrow C$. Complete tables of these factors $A_{2k}(\ )$ for practically all cases of nuclear radiation have been prepared by Biedenharn and Rose[40] in their extensive theoretical work on angular correlation of nuclear radiation.

A different, and perhaps more instructive, though less convenient, way to express angular correlation functions follows directly from (1), the $P_{2k}(\cos \Theta)$ being polynomials of $\cos \Theta$ of degree $2k$:

$$W_0(\Theta) = 1 + \sum_{k=1}^{k_{\max}} a_{2k} \cos^{2k}(\Theta). \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

In some cases it will be found appropriate to make use of the relationship $\cos^2 \Theta = \frac{1}{2}(1 + \cos 2\Theta)$ in order to express the angular correlation function in terms of an expansion in $\cos 2l\Theta$, rather than in terms of a power series in $\cos^2 \Theta$,

$$W_0(\Theta) = \sum_{l=0}^{l_{\max}} b_{2l} \cos 2l\Theta \quad . \quad . \quad . \quad . \quad . \quad . \quad (4\,a)$$

or more conveniently :

$$W_0(\Theta) = \mathrm{Re} \sum_{l=0}^{l_{\max}} b_{2l}\, \mathrm{e}^{2il\Theta} \quad . \quad . \quad . \quad . \quad . \quad . \quad (4\,b)$$

Simple expressions relate the coefficients a_{2k} and b_{2l} to the A_{2k} (Appendix I).

Angular correlation measurements became a practical possibility after the development of the scintillation counting method, which permits the measurement of nuclear radiation with considerable efficiency and speed. The first successful measurements were made by Brady and Deutsch[17] and the angular correlation method has since become one of the standard techniques of nuclear spectroscopy.

As an example, one of the most precisely measured angular correlations, the one of the ^{60}Ni gamma cascade[41], is shown in fig. 3. These gamma rays are emitted following the beta decay of ^{60}Co to ^{60}Ni. From experimental data other than correlation measurements, the three ^{60}Ni levels involved in this particular cascade are known to have angular momentum quantum numbers $I_A = 4$, $I_B = 2$ and $I_C = 0$, and the two gamma rays are both of the electric quadrupole type. For such a

gamma cascade, theory[29] predicts the following angular correlation function :

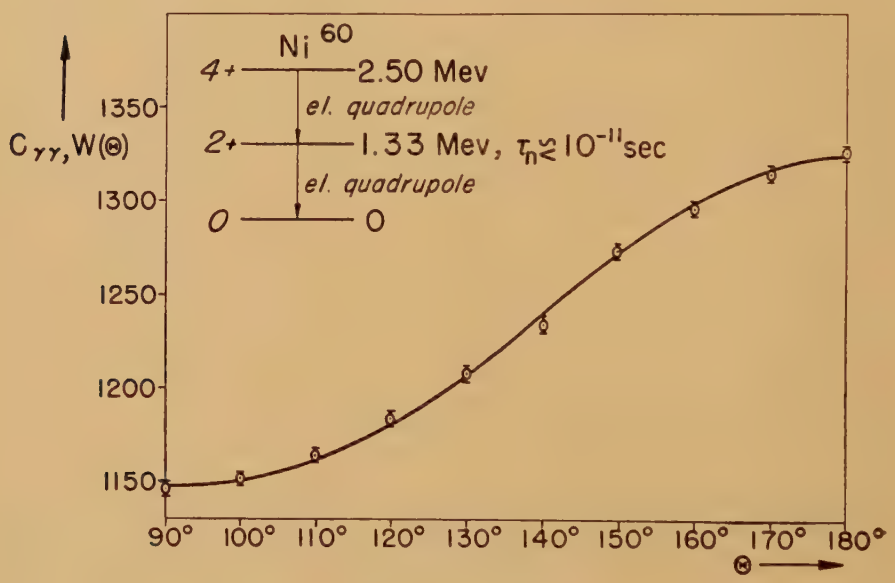
$$W_0(\Theta)=1+0{\cdot}102P_2(\cos\Theta)+0{\cdot}009P_4(\cos\Theta)$$

or

$$W_0(\Theta)=1+0{\cdot}1250\cos^2\Theta+0{\cdot}0417\cos^4\Theta.$$

This function, corrected for the finite solid angle subtended at the source by the two detectors, is represented by the solid line in fig. 3. The agreement of the experimental data and theory is excellent. This also indicates no observable disturbance of the angular correlation. In fact, a disturbance is hardly to be expected in this case, since during the very short lifetime of the intermediate ^{60}Ni state, approximately 10^{-11} sec, the probability of transitions between the magnetic substates is extremely small, at least if we exclude the possibility of very special conditions in the source (e.g. large anisotropic hyperfine structure as in Co–Tutton salts).

Fig. 3



The angular correlation of the ^{60}Ni gamma rays.

A convenient quantity to use in discussing angular correlation experiments is the '*anisotropy value*'

$$A=\frac{W(180°)-W(90°)}{W(90°)}=\frac{W(180°)}{W(90°)}-1 \quad . \ . \ . \ . \quad (5)$$

which characterizes the anisotropic behaviour of the correlation at 180° relative to 90°. A zero value of A means in most (not all) cases that the correlation is isotropic. One advantage of the $\cos^2\Theta$ expansion (3)

is the simple relationship between the anisotropy A and the coefficients a_{2k}:

$$A = \sum_{k=1}^{k_{\max}} a_{2k}. \qquad (6)$$

In the case of the ^{60}Ni gamma cascade the theoretical value of A is $+0{\cdot}16667$, the experimental $+0{\cdot}167 \pm 0{\cdot}001$.

§ 3. The Interaction of Nuclear Moments with External Fields

3.1. *The Nuclear Moments*

Apart from the angular momentum $\mathbf{I}\hbar$, nuclear states also possess a magnetic dipole moment $\boldsymbol{\mu}$ and an electric quadrupole moment Q. The latter measures the deviation of the nuclear charge distribution from spherical symmetry. A positive value of Q arises from a homogeneous charge distribution elongated with respect to the nuclear axis. A flattened homogeneously charged nucleus has a negative quadrupole moment. Electric dipole moments are generally assumed not to exist in nuclei. Higher multipoles than the ones mentioned may, in principle, be present, but their possible effects can be neglected here. If we place such a nucleus in an external magnetic or electric field it interacts with these fields by virtue of its magnetic and electric moments.

3.2. *Static Magnetic Field*

A static magnetic field $\mathbf{H}$ exerts a torque on the nuclear magnetic dipole $\boldsymbol{\mu}$ and tends to align the nuclear magnetic axis in the direction of $\mathbf{H}$. As expected and as demonstrated in nuclear magnetic resonance experiments, the nuclear angular momentum $\mathbf{I}\hbar$ (whose direction coincides with the magnetic axis of the nucleus) will respond to this torque by a precession around $\mathbf{H}$ as axis (fig. 4). A simple classical calculation (cf. ref. (24)) yields for the angular precession velocity (Larmor-frequency):

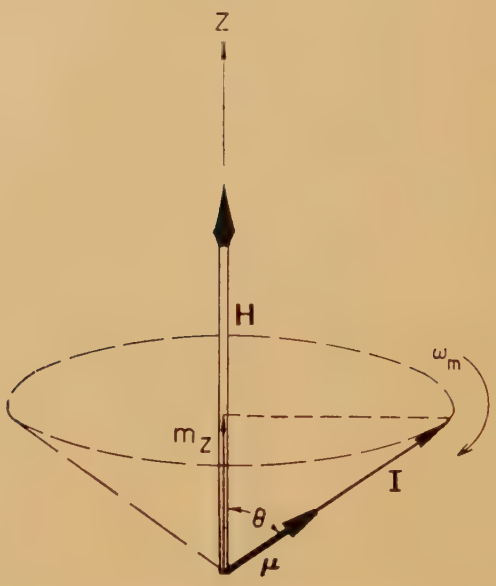
$$\omega_m = \frac{\mu H}{I\hbar}. \qquad (7)$$

The precession proceeds in a definite sense determined by the sign of μ. In addition, the Larmor-frequency ω_m is independent of the relative orientation of $\boldsymbol{\mu}$ and $\mathbf{H}$.

Quantum mechanics enters the picture in so far as it restricts the angles θ between the axis of $\boldsymbol{\mu}$ and the field $\mathbf{H}$ to those which result in a component of $\mathbf{I}$ along $\mathbf{H}$ equal to an integer m_z (magnetic quantum number) $I \geqslant m_z \geqslant -I$, thus allowing $2I+1$ different orientations of $\mathbf{I}$ with respect to $\mathbf{H}$. Since the potential energy of a magnetic dipole in a magnetic field depends upon the relative orientation:

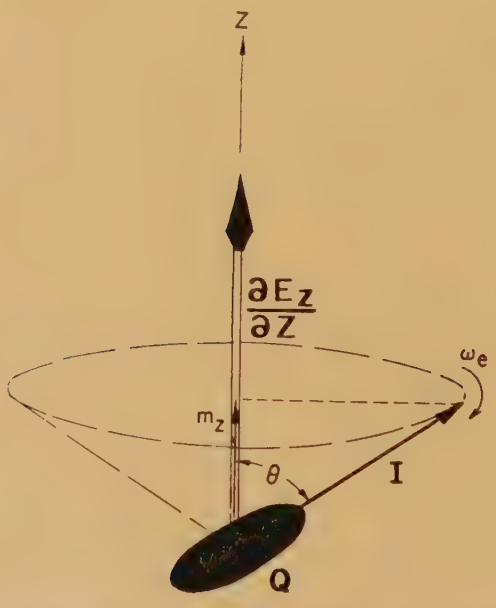
$$E_m(m_z) = -\boldsymbol{\mu} \cdot \mathbf{H} = -\frac{\mu \cdot H}{I} m_z$$

Fig. 4



The precession of the nuclear angular momentum **I** and the magnetic moment μ around a magnetic field **H**.

Fig. 5



The precession of the nuclear angular momentum **I** and the electric quadrupole moment Q around the symmetry axis of an electrostatic gradient.

the $2I+1$ substates differ in their energy values. The magnetic splitting, the energy difference between two adjacent levels ($\Delta m_z = \pm 1$), is constant

$$\Delta E_m = \frac{\mu H}{I} = \omega_m \hbar$$

and when expressed in terms of the frequency $\Delta E_m/\hbar$, corresponds to the Larmor-frequency of the precessing magnetic dipole.

In a similar way, the energy splitting due to the presence of the nuclear magnetic moment in the magnetic field of the shell electrons gives rise to the well-known magnetic hyperfine structure of spectral lines.

Nuclear magnetic moments as large as $+5{\cdot}5$ nuclear magnetons have been observed[42]. The frequency ω_m of the Larmor precession of such a nucleus in a magnetic field of $H = 10\,000$ gauss is about $10^7\ \mathrm{sec}^{-1}$.

3.3. *Static Gradient of an Electric Field*

The situation is slightly more complicated in the case of the electric interaction. Since there is no nuclear electric dipole moment, no torque is exerted on the nucleus in a homogeneous electric field. An interaction with the electric quadrupole moment of the nucleus necessitates a gradient of the electric field : Grad **E**. The latter is a second order tensor quantity requiring, in general, nine components for its specification. By a proper choice of axes (transformation to principal axes) and by virtue of certain properties of the electric field (Laplace's law) the number of components can be reduced to two, and further, if we assume axial symmetry of the field (e.g. around the z-axis), then the gradient is given by one number alone, e.g. $\partial E_z/\partial z$ or $\partial^2 V/\partial z^2$. The generalization to non-axial (rhombic) fields, however, can be made easily by introducing an asymmetry parameter

$$\epsilon = \left| \frac{\partial E_x}{\partial x} - \frac{\partial E_y}{\partial y} \right| \Big/ \left| \frac{\partial E_z}{\partial z} \right| .$$

For simplicity axially symmetric electric gradients are assumed in the immediate discussion.

The interaction between such an electric gradient and a nuclear electric quadruple moment gives rise to an aligning torque exerted on the nucleus. The resulting ' precession ' of the angular momentum about the axis z of the gradient field has in general *not one*, but *several* characteristic frequencies, depending upon the relative orientation of the nuclear spin axis **I** with respect to the axis of the field (z-axis) (fig. 5). This is also reflected in the non-equidistant splitting of the $2I+1$ energy levels caused by the electric quadrupole coupling, a fact which is well known from pure quadrupole spectra[43]. The potential energy of a quadrupole system, where the position of the axis of the nuclear quadrupole moment with

respect to the field axis (z-axis) is specified by the magnetic quantum number m_z (component of **I** along the symmetry (z)-axis), is given by

$$E_Q(m_z)=\frac{3m_z^2-I(I+1)}{4I(2I-1)}eQ\frac{\partial E_z}{\partial z}. \quad . \quad . \quad . \quad . \quad . \quad (8)$$

Positions of **I** corresponding to $+m$ and $-m$ (angle θ and $180^\circ-\theta$) have the same energy; the levels are twofold degenerate. A classical interpretation of this energy degeneracy would make use of the following picture: The quadrupole nucleus (actually the vector **I**) precesses in one direction for the angle $\theta<90^\circ$ $(+m)$ and with the same precession frequency but in the opposite direction for the angle $180^\circ-\theta$ $(-m)$. Hence, the quadrupole precession is not unidirectional like the magnetic precession. Again the characteristic frequencies, which we can visualize classically as the precession frequencies of the quadrupole moment around the field axis, correspond to the energy differences between neighbouring levels, expressed in terms of frequency

$$\omega_e=\frac{\Delta E_Q}{\hbar}=\frac{E_Q(m_z)-E_Q(m_z')}{\hbar}. \quad . \quad . \quad . \quad . \quad . \quad (9)$$

Table 1. Characteristic 'Precession' Frequencies for Electric Quadrupole Interaction

Angular momentum quantum number of the nucleus I	Characteristic interaction frequencies ω_e in multiples of the fundamental frequency $\omega_e{}^0$
1	1
3/2	1
2	1, –, 3, 4
5/2	1, 2, 3
3	1, –, 3, 4, 5, –, –, 8, 9

For a few values of I the possible angular frequencies are given in table 1 in terms of the fundamental 'precession' frequency, the frequency equivalent of the smallest non-vanishing energy difference which is

$$\omega_e{}^0=\frac{3}{4I(2I-1)}\frac{eQ}{\hbar}\cdot\frac{\partial E_z}{\partial z} \text{ for integer } I, \quad . \quad . \quad . \quad (10\,a)$$

$$\omega_e{}^0=\frac{3}{2I(2I-1)}\frac{eQ}{\hbar}\frac{\partial E_z}{\partial z} \text{ for half integer } I. \quad . \quad . \quad (10\,b)$$

Inasmuch as quadrupole interaction frequencies ω_e as large as $2\times10^{10}\,\mathrm{sec}^{-1}$ have been observed[44] (^{127}I in ICl molecule) the electrostatic quadrupole interaction is by no means a minor one.

3.4. *Static Interactions and the Angular Correlation Process (Qualitative Considerations)*

So far we have considered static extranuclear fields of constant magnitude and direction. The common feature of these static interactions is the occurrence of a precession of the nuclear spin around a well-defined, stationary axis, the symmetry axis of the field. The latter was also used as the axis of quantization for which the m_z values of the angular momentum projection were defined. By the precession of I these projections do not change ; the interaction mechanisms do not induce transitions between the m_z-sublevels, defined with respect to the *field axis as the axis of quantization*. Nevertheless, certain phase relationships between the m_z-sublevels are introduced by the precession of the angular momentum. The population of m-sublevels, however, defined with respect to a direction other than the field direction as the axis of quantization, is changing periodically with the precession of I about the field axis. The influence of such a precessing nucleus on the angular correlation is now evident.

(i) *The parallel field case.* If the field axis z coincides with the axis of quantization $\mathbf{k}_1$ which was used in the discussion of the angular correlation for the introduction of the unequally populated m_{k_1} states, then the m_z are the same as the m_{k_1}. Since the m_z population does not change, the m_{k_1} population is not affected either ; the angular correlation remains unperturbed. It is even possible to apply a strong external magnetic field in the direction of $\mathbf{k}_1$ and thus ' decouple ' any other static magnetic interaction which might be present and restore the full unperturbed angular correlation : the exact analogue to the Paschen–Back effect in optical spectroscopy.

(ii) *The general case of the static field.* If the field axis z has a direction different from the axis of quantization $\mathbf{k}_1$, then $m_z \neq m_{k_1}$, the population of the m_{k_1} changes due to the continually (but periodically) changing phase in the m_z states, which, however, does not affect the population of the m_z states at all. Since for the angular distribution of the second radiation the m_{k_1} population is relevant, the presence of such a non-parallel field alters the angular correlation. In general, however, the periodic change of the m_{k_1} population by the precession of the nucleus around z obviously does not lead to an *equal* population of the m_{k_1} substates, thus a complete destruction of the angular correlation is not expected from such a static interaction ; a so-called ' hard core ' correlation remains. An exception is the case of the ' perpendicular field ' (see below). It is interesting to note that in case the precession frequencies are a multiple of a ' fundamental ' frequency ω^0 (true for

external magnetic field without I–J coupling and axially symmetric electric gradient) a measurement of the second nuclear radiation, after a time, equal to the fundamental precession period $T_0=2\pi/\omega^0$ has elapsed, would exhibit again the unperturbed correlation.

(iii) *The perpendicular field case.* A complete disappearance of the angular correlation in the case of a static interaction is also possible if (*a*) the symmetry axis of the extranuclear field (z-axis) is perpendicular to both directions of emission of nuclear radiation and (*b*) if the directions $+z$ and $-z$ cannot be distinguished with reference to the two directions $\mathbf{k}_1$ and $\mathbf{k}_2$. The condition (*b*) is fulfilled if the two counters cannot distinguish between the two radiations of the nuclear cascade or if the field acting on one half of the nuclei in the source is directed toward $+z$ and on the other half toward $-z$.

3.5. *Time-dependent Fields*

Time-dependent interaction mechanisms due to electric and magnetic fields, randomly fluctuating in direction, induce transitions between the different m-states defined with respect to *any* axis of quantization. Ultimately this is a consequence of the fact that this interaction mechanism by its randomness does not introduce any preferred direction at the site of the nucleus, and thus does not specify an axis of quantization by itself. It is the same mechanism which brings the nuclear spin system in nuclear magnetic resonance absorption back to thermal equilibrium with its environment, and thus is responsible for the spin-lattice relaxation time T_1. Provided there is enough time available, this relaxation mechanism results in the situation that a nucleus of angular momentum I can be found in *any* m-substate with *equal probability*, whatever the initial population of the m-states. The ' memory ' of the presence of any preferred direction introduced by some earlier process may become completely lost. The approach to uniform population is exponential: $e^{-\lambda t}$, the corresponding relaxation constant λ being the analogue of the inverse of the spin-lattice relaxation time T_1.

Again the effect on the angular correlation process is obvious[45,46]: The longer the time t, during which the intermediate nuclear state finds itself subjected to the time-dependent interaction, the more isotropic the correlation. A complete ' loss of memory ' results in a perfectly isotropic angular distribution of the radiation.

3.6. *Radiofrequency Fields—Nuclear Resonance Transitions*

Transitions between m-substates, differing in energy by ΔE due to some static external field, can also be induced by an oscillating electromagnetic field of the corresponding frequency $\omega_R=\Delta E/\hbar$[46], and the resulting change in the angular correlation could serve as an indicator of the occurrence of the resonance transitions. The possibility of very

accurate frequency determination hence could be employed to obtain precise values of the energy splitting of the m-states and thus would allow a precision measurement of the nuclear moments involved in the intermediate state.

The radiofrequency fields, however, must be of such an amplitude that sufficient resonance transitions are induced within the lifetime of the nuclear state, to give an observable effect on the angular correlation. This requires extremely strong radiofrequency fields, which makes such an experiment hardly feasible, at least at present.

§ 4. Theory of the Influence of Extranuclear Fields on the Angular Correlation

4.1. *General Considerations*

Of fundamental importance for the occurrence of an anisotropic angular correlation is the unequal population of the $2I_B+1$ magnetic sublevels in the intermediate nuclear state. In the absence of any external field the sublevels corresponding to different orientations of the nuclear spin all have the same energy. The level is said to be $2I_B+1$ fold degenerate. In this case no transitions between these levels take place and the unperturbed angular correlation is observed.

The case of a vanishing extranuclear field is of somewhat academic interest, since, in reality, external fields do exist in most cases as a result of the charge distribution of the electron shell and other atoms surrounding the decaying nucleus.

Frequently no coupling is to be expected between the nucleus and the shell for a free atom (e.g. atom in S_0 state). However, one must keep in mind that the atom of the decaying nucleus is in most cases a constituent of a solid or a liquid, where the electronic configuration of a given atom is in general quite different from the ground state of the electron shell of the *free* atom. Furthermore, as a result of preceding nuclear decay processes, the shell may not be in the ground state during the time the nuclear cascade proceeds.

Time-dependent fields are also present: in solids, due to lattice vibrations and internal motions of atoms; in liquids, due to the constant change of local configurations resulting from the Brownian motion.

An observable influence of static and time-dependent extranuclear fields on the angular correlation is to be expected only when an appreciable change of the m_{k_1}-population (defined with respect to the propagation direction $\mathbf{k}_1$ of the first radiation) is induced. For this to happen requires not only the presence of perturbing fields but also sufficient time t for the m_{k_1}-transitions to proceed. For the static interaction, where we can define a ' precession ' frequency ω, the relative attenuation of the angular correlation function is of the order $(\omega t)^2$ (for $\omega t<1$) and it is of the order λt for time-dependent interaction mechanisms. Present experimental techniques allow, under favourable circumstances, the

detection of attenuation effects which reduce the anisotropy by not more than a few per cent. Thus the necessary condition for an observable influence of a static field on a nuclear angular correlation is $\omega t < 0{\cdot}1$ and similarly, for time-dependent fields, $\lambda t < 0{\cdot}05$.

Quantitatively it can be stated that only in extraordinary cases are intermediate states of lifetimes shorter than 10^{-10} sec affected by extranuclear fields. As a matter of fact the majority of nuclear gamma transitions have appreciably shorter lifetimes.

How is the angular correlation function of a given nuclear cascade quantitatively altered by the interactions in the intermediate state? The time interval t, during which the intermediate nuclear state is exposed to the interacting fields, is essentially determined by the nuclear lifetime τ_N, but, in addition, depends also upon the time characteristics of the coincidence analyser used to measure the angular correlation.

Assume that the first radiation of the nuclear cascade is observed at the time $t=0$ and the second radiation is accepted only between t and $t+dt$. Then the angular correlation $W(\Theta)$ will also be a function of the 'delay time' t, the nuclear precession having this time available to proceed. The corresponding angular correlation $W(\Theta, t)$ is called *delayed correlation*. For an actual experiment, of course, one must take into account the finite resolving time τ_0 of the coincidence analyser, which records the second radiation between $t-\tau_0$ and $t+\tau_0$ seconds after the emission of the first.

Most angular correlation measurements are performed without using an artificial time delay introduced by the measuring apparatus. The second nuclear radiation is then observed within the resolving time τ_0 of the coincidence analyser, from $t=0$ to $t=\tau_0$. Then the correlation function becomes

$$W_{\tau_0}(\Theta)=\frac{\int_0^{\tau_0} \exp(-t/\tau_N) \cdot W(\Theta, t)\, dt}{\int_0^{\tau_0} \exp(-t/\tau_N) \cdot dt} \quad . \quad . \quad . \quad . \quad . \quad (11)$$

For infinite resolving time ($\tau_0 \gg \tau_N$), the so-called *integral* or *average* correlation is observed:

$$W_{\infty}(\Theta)=\frac{1}{\tau_N}\int_0^{\infty} \exp(-t/\tau_N) \cdot W(\Theta, t)\, dt, \quad . \quad . \quad . \quad (12)$$

which is now independent of apparative constants.

Two cases of interacting fields must be distinguished.

(i) *The perturbing interaction does not introduce a preferred direction in the radioactive source as a whole.* This condition is satisfied for an isotropic radioactive source which is not subjected to an external field. Since most radioactive sources are in the form of a polycrystalline powder or an isotropic liquid, this is the most frequently encountered case.

The ' isotropic ' interaction does not result in a complete change of the angular correlation pattern ; it only reduces the anisotropic behaviour of the correlation. The correlation function retains its essential form (1) but each correlation coefficient A_{2k} is multiplied by an attenuation factor G_{2k} $(G_{2k} \leqslant 1)$[47, 48] which is characteristic of the perturbing interaction

$$W(\Theta) = 1 + \sum_{k=1}^{k_{\max}} G_{2k} A_{2k} P_{2k} (\cos \Theta). \quad . \quad . \quad . \quad . \quad (13)$$

The G_{2k} depend *only* upon the parameters describing the interaction in the intermediate nuclear state and upon the time characteristics of the experimental arrangement. They are independent of the type of nuclear radiations and of the characteristics of the initial and final states involved in the nuclear cascade. For the *static* ' isotropic ' interaction the G_{2k} will never vanish completely[46, 48] ; a limiting or ' hard core ' correlation remains. In fact it can be shown that the irreducible minimum of G_{2k} for the integral correlation and for axially symmetric static interaction is[46, 48]

$$G_{2k}(\min) = \frac{1}{4k+1}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (14)$$

In contradistinction, time-dependent interacting fields, as outlined above, can wipe out completely the anisotropic correlation, i.e. G_{2k} can become vanishingly small[45, 46].

(ii) *The perturbing interaction gives rise to the existence of a privileged direction in the radioactive source.* Externally applied fields or internal fields in an anisotropic crystalline source may be responsible for the existence of a preferred axis at the source. The angular correlation will then depend upon the relative directions of the emission of the two nuclear cascade radiations $\mathbf{k}_1$ and $\mathbf{k}_2$, measured with respect to the preferred axis, which we let coincide with the z-axis of a coordinate system xyz (fig. 6). The mathematical expression describing such an angular correlation will thus involve functions which are not simply dependent upon the *angle* Θ between $\mathbf{k}_1$ and $\mathbf{k}_2$, like the Legendre polynomials, but also functions of the $\mathbf{k}_1$, $\mathbf{k}_2$ directions in space with respect to the z-axis. Such functions are the spherical harmonics $Y_k^l(\theta, \phi)$. The general form of the angular correlation function is now rather complicated[46] :

$$W(\theta_1\phi_1\theta_2\phi_2) = \mathrm{Re} \sum_{k_1k_2l_1l_2} I_{k_1} II_{k_2} III_{k_1k_2}^{l_1l_2} Y_{k_1}^{l_1}(\theta_1\phi_1) Y_{k_2}^{l_2}(\theta_2\phi_2). \quad . \quad (15)$$

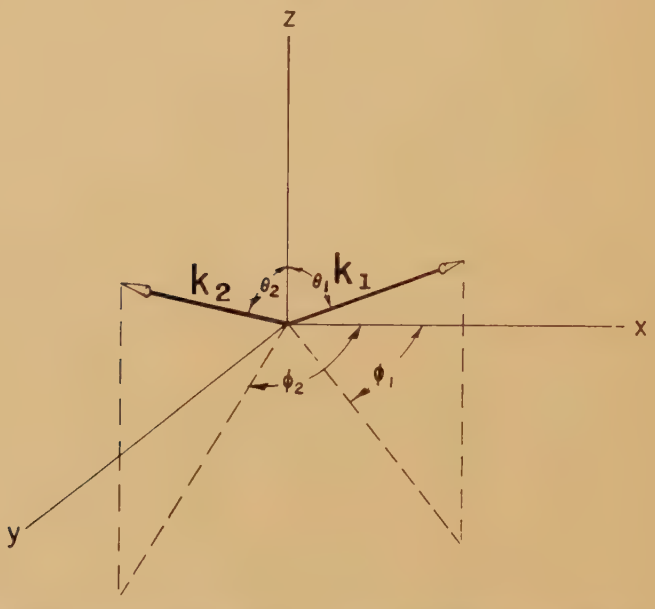
Again, all the parameters contained in I_{k_1} (II_{k_2}) refer to the first (second) nuclear transition only, whereas the attenuation coefficients $III_{k_1k_2}^{l_1l_2}$ are functions of the interaction parameters in the intermediate state and like G_{2k} depend upon the time characteristics of the coincidence analyser. Special cases of expression (15) will be discussed later, when we consider specific examples of interactions in anisotropic sources (see also Appendix II).

4.2. *Static Magnetic Interaction between the Electron Shell and the Nucleus*

4.2.1. *Isotropic Hyperfine Coupling in Free Atoms*

The coupling between the magnetic moment μ of a nucleus and the magnetic field due to the electronic currents in the atomic shell is well known from the magnetic hyperfine structure of spectral lines. The influence of this I–J coupling on the angular correlation was first considered in a theoretical paper by Goertzel[49], who assumed J to be a constant of the motion. This theory thus holds for free atoms in a stationary state, an important restriction, which is seldom satisfied in

Fig. 6



The coordinates for the specification of the propagation directions $\mathbf{k}_1$ and $\mathbf{k}_2$

practical cases. Numerical calculations for the special, but representative, case $J=\frac{1}{2}$ were performed by Alder[48] with the following result for the attenuation coefficients (integral correlation):

$$G_{2k}(\omega_m\tau_N)_\alpha = 1 - \frac{2k(2k+1)}{(2I_B+1)^2}\frac{(\omega_m\tau_N)^2}{1+(\omega_m\tau_N)^2} \quad (J=\tfrac{1}{2}). \tag{16}$$

ω_m is the Larmor frequency of the precession of $\mathbf{I}_B$ around $\mathbf{F}=\mathbf{I}_B+\mathbf{J}$. For larger values of $\omega_m\tau_N$ (strong I–J coupling) the limiting values

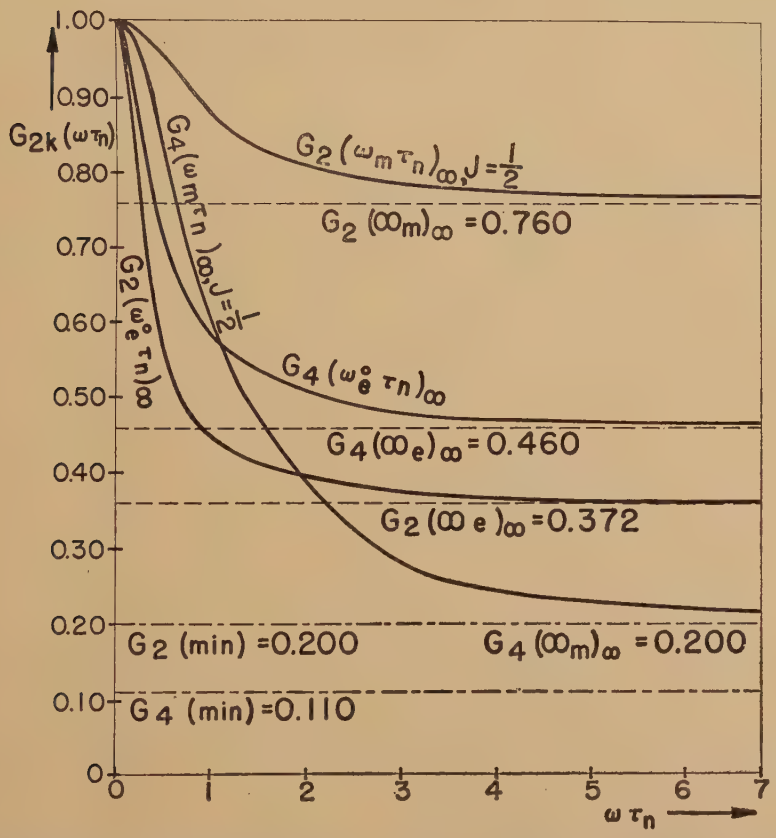
$$G_{2k}(\infty)_\alpha = 1 - \frac{2k(2k+1)}{(2I_B+1)^2} \quad . \quad . \quad . \quad . \quad . \tag{17}$$

which, in general, are larger than the irreducible minima $G_{2k}(\min)$ for *any* static interaction with axial symmetry (eqn. (14)) are approached asymptotically (fig. 7).

4.2.2. *Effect of an External Magnetic Decoupling Fiel* (40, 49)

If an external magnetic field H_0 is applied, the vector resultant **F** of the electron shell angular momentum **J** and the nuclear momentum $\mathbf{I}_B$ will precess about the field axis (fig. 8(*a*)), while $\mathbf{I}_B$ and **J** both precess about **F** (Zeeman effect of the hyperfine structure). The magnetic moment μ_J associated with the J of the electron shell is in general about a thousand times larger than the nuclear moment μ_I, and thus the

Fig. 7



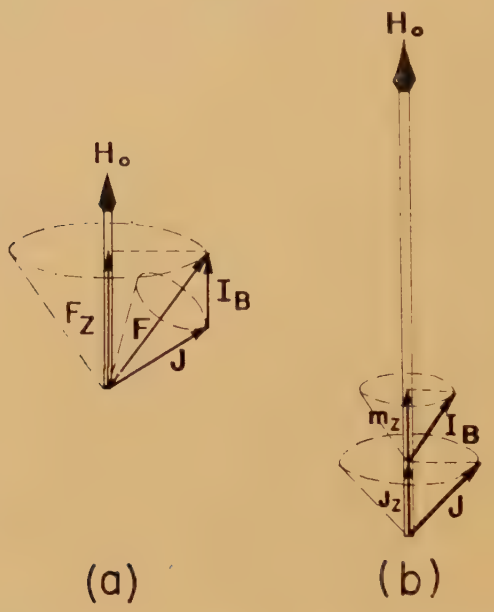
The attenuation coefficients $G_2(\omega\tau_N)_\infty$ and $G_4(\omega\tau_N)_\infty$ for static magnetic interaction (I–J coupling) and static electric quadrupole interaction (I_B=2).

interaction of H_0 with μ_J is as many times larger than the interaction between H_0 and μ_I. In fact, for a large enough field the interaction energy between H_0 and μ_J can become much larger than the interaction between μ_I and μ_J, and this strong interaction with the external field causes a breaking of the I–J coupling. The electron shell **J** and the nuclear magnetic moment $\boldsymbol{\mu}_I$ both precess independently in well-defined positions about $\mathbf{H}_0$ (fig. 8(*b*)). The nuclear angular momentum **I** is

completely decoupled from the electron shell (complete Paschen–Back effect of the hyperfine structure). The effect on the angular correlation is now entirely due to the interaction of the nuclear magnetic moment with the external field H_0.

If the strong external field H_0 is applied in the direction of the emission of one nuclear radiation, we have the 'parallel field case', and the correlation is the same as if no magnetic interaction were present.

Fig. 8



The Zeeman effect of hyperfine structure (*a*) and the decoupling of **I**–**J** in a strong external magnetic field (*b*).

Thus the full, unperturbed angular correlation is observed. The condition for $\mathbf{H}_0$ to restore an angular correlation otherwise attenuated by the magnetic I–J coupling, is[49]

$$H_0^2 \gg (2\times 10^4 \Delta\nu), \qquad \ldots\ldots (18)$$

where $\Delta\nu$ is the overall hyperfine splitting in cm^{-1} and H_0 is in gauss. Judging from optically determined values of $\Delta\nu$ a field of about 10000 gauss should be adequate in most cases. The possibility of a magnetic decoupling was first pointed out by Goertzel[49].

4.2.3. *Static Anisotropic Hyperfine Coupling in Solids*[46]

The hyperfine structure in free atoms is isotropic due to the random orientation of **J**. In solids, however, there usually exists a preferred direction of **J** as a result of the crystalline structure. This anisotropy

is particularly pronounced in the paramagnetic atoms of the iron group[50,51], where it has been studied extensively both by theoreticians and experimentalists. Crystals with large anisotropies of their hyperfine structure are used, for instance, in Bleaney's method for aligning nuclear angular momenta at low temperatures[52].

The effect of the anisotropic magnetic hyperfine coupling on the intermediate nuclear state has been considered by Abragam and Pound[46] in their extensive theoretical investigation of the influence of electric and magnetic fields on angular correlations. In contradistinction to the isotropic *I–J* coupling in free atoms, an external magnetic decoupling field does not restore the full angular correlation in polycrystalline sources which show a large anisotropy of the magnetic hyperfine structure.

4.3. *Static External Magnetic Field, no I–J Coupling*

Calculations of the influence of an external magnetic field on the angular correlation of nuclear radiation, neglecting electron shell interactions, have been made by Alder[48,53] and Lloyd[54]. For equidistant magnetic hyperfine splitting the general expression (15) for the angular correlation with preferred direction interactions admits of considerable simplification[48] (cf. Appendix II):

$$W(\theta_1\phi_1, \theta_2\phi_2)=\sum_{k,l} g^{(l)} I_{2k} II_{2k} Y_{2k}^{\,l}(\theta_1\phi_1) Y_{2k}^{\,-l}(\theta_2\phi_2), \quad . \quad . \quad (19)$$

where the attenuation coefficients of the delayed correlation are independent of the index $2k$:

$$g^{(l)}(t)=\exp(il\omega_m t). \quad . \quad . \quad . \quad . \quad . \quad . \quad (20)$$

For the time-integrated correlations the attenuation coefficients are easily obtained from (20) (compare eqns. (11) and (12)):

$$g^{(l)}(\omega_m\tau_N)_{\tau_0}=\frac{1}{1-il\omega_m\tau_N}\,\frac{1-\exp(il\omega_m\tau_0)\,.\,\exp(-\tau_0/\tau_N)}{1-\exp(-\tau_0/\tau_N)} \quad (21\,a)$$

and for $\tau_0 \gg \tau_N$

$$g^{(l)}(\omega_m\tau_N)_{\infty}=\frac{1}{1-il\omega_m\tau_N}=\frac{1+il\omega_m\tau_N}{1+(l\omega_m\tau_N)^2}. \quad . \quad . \quad . \quad . \quad . \quad (21\,b)$$

Especially simple and useful is the case of the external magnetic field applied perpendicular to the directions of emission of the two nuclear radiations. For symmetry reasons the angular correlation becomes again a function of the angle $\Theta=\phi_1-\phi_2$ between $\mathbf{k}_1$ and $\mathbf{k}_2$ only:

$$W(\Theta, t)=\sum_{l=0}^{l_{\max}} g^{(2l)}(t)\,.\,b_{2l}\exp(i2l\Theta), \quad . \quad . \quad . \quad . \quad (22)$$

where the b_{2l} are the coefficients of the unperturbed correlation (4 *a*).

The delayed correlation

$$W(\Theta, t)=\sum_{l=0}^{l_{\max}} b_{2l}\exp[i2l(\Theta+\omega_m t)] \quad . \quad . \quad . \quad . \quad (23)$$

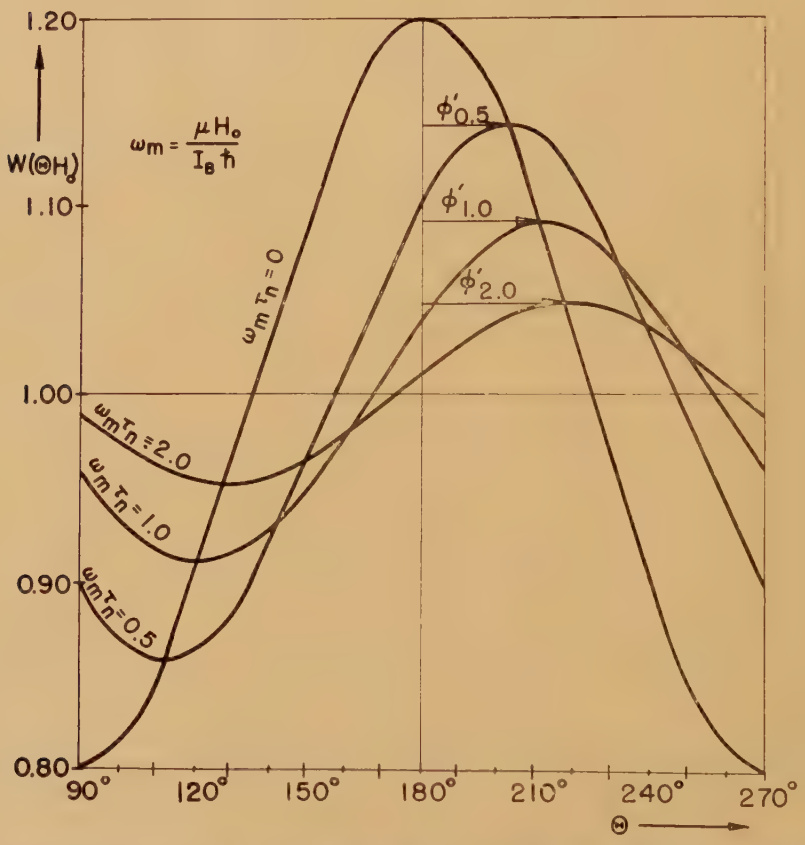
differs from the unperturbed correlation (4 *a*) only by the azimuthal shift $\omega_m t$. As far as the integral correlation is concerned

$$W_\infty(\Theta)=\mathrm{Re}\sum_{l=0}^{l_{\max}}\frac{1+i2l\omega_m\tau_N}{1+(2l\omega_m\tau_N)^2}\cdot b_2\exp(i2l\Theta)$$

$$=\sum_{l=0}^{l_{\max}}\frac{b_{2l}}{1+(2l\omega_m\tau_N)^2}[\cos 2l\Theta-2l\omega_m\tau_N\sin 2l\Theta), \quad . \ . \ . \quad (24)$$

the magnetic field $\mathbf{H}_0$ causes an attenuation in addition to the azimuthal shift (fig. 9). Again, but approximately correct only if $\omega_m\tau_N<1$, the correlation pattern is rotated by the classical precession angle $\phi=\omega_m\tau_N$.

Fig. 9



The attenuation and 'azimuthal shift' ϕ of an angular correlation ($W_0(\Theta, 0)=1+0{\cdot}2\cos 2\Theta$) in a magnetic field H_0 perpendicular to the plane ($\mathbf{k}_1, \mathbf{k}_2$).

A determination of the attenuation and the direction of the azimuthal shift for a given value of $\mathbf{H}_0$ yields magnitude and sign of $\omega_m\tau_N$. Knowing the nuclear lifetime τ_N, the magnetic moment μ_B of the intermediate

nuclear state can be determined in size and sign. So far such a measurement, first proposed by Goertzel[49], has been accomplished in two cases (see Part II).

If the two detectors used to measure the angular correlation are equally sensitive for the two radiations $g^{(2l)}$ becomes real:

$$G^{(2l)}(\omega_m\tau_N)_\infty = \frac{1}{2}\left[\frac{1+i2l\omega_m\tau_N}{1+(2l\omega_m\tau_N)^2}+\frac{1-i2l\omega_m\tau_N}{1+(2l\omega_m\tau_N)^2}\right]=\frac{1}{1+(2l\omega_m\tau_N)^2} \quad (25)$$

and the correlation function is symmetric with respect to $\Theta=180°$:

$$\overline{W}_\infty(\Theta)=\sum_{l=0}^{l_{\max}}\frac{1}{1+(2l\omega_m\tau_N)^2}b_{2l}\cdot\cos(2l\Theta). \quad . \quad . \quad . \quad (26)$$

4.4. *Static Electric Quadrupole Interaction*

4.4.1. *Crystalline Powder (Isotropic Sources)*

The existence of a strong interaction of nuclear electric quadrupole moments with electric field gradients in crystals and in molecules is well known from nuclear magnetic resonance absorption and from pure quadrupole spectra. For axially symmetric fields (hexagonal, tetragonal crystal structure) and for isotropic sources (e.g. composed of a large number of randomly orientated microcrystals) the attenuation coefficients $G_{2k}(t)$ for the delayed correlation have been calculated by Abragam and Pound[46]:

$$I_B=1: \quad G_2(t)=\tfrac{1}{5}(3+2\cos\omega_e{}^0t), \quad . \quad . \quad . \quad (27)$$

$$I_B=\tfrac{3}{2}: \quad G_2(t)=\tfrac{1}{5}(1+4\cos\omega_e{}^0t), \quad . \quad . \quad . \quad (28)$$

$$I_B=2:\begin{cases} G_2(t)=\tfrac{1}{35}(13+2\cos\omega_e{}^0t+12\cos 3\omega_e{}^0t+8\cos 4\omega_e{}^0t), & (29\,a)\\ G_4(t)=\tfrac{1}{63}(29+12\cos\omega_e{}^0t+16\cos 3\omega_e{}^0t+6\cos 4\omega_e{}^0t), & (29\,b)\end{cases}$$

$$I_B=\tfrac{5}{2}:\begin{cases} G_2(t)=\tfrac{1}{35}(7+13\cos\omega_e{}^0t+10\cos 2\omega_e{}^0t+5\cos 3\omega_e{}^0t), & (30\,a)\\ G_4(t)=\tfrac{1}{63}(7+15\cos\omega_e{}^0t+18\cos 2\omega_e{}^0t+23\cos 3\omega_e{}^0t), & (30\,b)\end{cases}$$

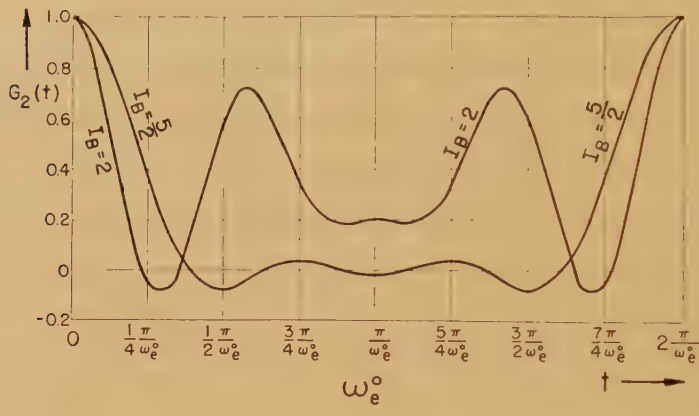
$$I_B=3:\begin{cases} G_2(t)=\tfrac{1}{105}(33+2\cos\omega_e{}^0t+15\cos 3\omega_e{}^0t+20\cos 4\omega_e{}^0t \\ \qquad\qquad +25\cos 5\omega_e{}^0t+10\cos 8\omega_e{}^0t), & (31\,a)\\ G_4(t)=\tfrac{1}{693}(187+30\cos\omega_e{}^0t+92\cos 3\omega_e{}^0t+6\cos 4\omega_e{}^0t \\ \qquad\qquad +60\cos 5\omega_e{}^0t+192\cos 8\omega_e{}^0t+126\cos 9\omega_e{}^0t), & (31\,b)\\ G_6(t)=\tfrac{1}{1\,29}(165+50\cos\omega_e{}^0t+69\cos 3\omega_e{}^0t+32\cos 4\omega_e{}^0t \\ \qquad\qquad +67\cos 5\omega_e{}^0t+34\cos 8\omega_e{}^0t+12\cos 9\omega_e{}^0t), & (31\,c)\end{cases}$$

$\omega_e{}^0$ is the fundamental ' precession ' frequency (10) of the electric quadrupole moment. We recognize in the arguments of the cos functions the characteristic frequencies of table 1 corresponding to the different I_B

values. In fig. 10 the coefficients $G_2(t)$ for $I_B=2$ and $I_B=5/2$ are plotted as a function of the delay time t. It is interesting to note that the attenuation coefficients for the delayed correlation can take values lower than the minimum values $G_{2k}(\min)$. In fact they even can become negative (fig. 10).

The determination of $G_{2k}(t)$ by means of delayed coincidence techniques allows the measurements of $\omega_c{}^0$ for polycrystalline sources. Such an experiment, however, is possible only when the following conditions are fulfilled. To extend the coincidence measurements to a delay time of $t=T_0=2\pi/\omega_c{}^0$ (the quadrupole precession period), the nuclear lifetime τ_N must not be much smaller than T_0. Secondly, due to the finite resolving time τ_0 of the coincidence analyser, $G_{2k}(t)$ will be smeared out. To keep this effect within tolerable limits, τ_0 must be appreciably smaller than T_0.

Fig. 10



The attenuation coefficient $G_2(t)$ for the delayed correlation as a function of the delay time t for $I_B=\frac{5}{2}$ and $I_B=2$ (according to Abragam and Pound, reference (46)).

Once $\omega_c{}^0$ is known the magnitude of the electric quadrupole moment (but not its sign) could be determined from (10) if the gradient of the crystalline electric field $\partial E_z/\partial z$ were known at the lattice site of the decaying nucleus. A reliable calculation of this quantity is difficult under normal circumstances and is made even more complex in this case since the environment of the nucleus in the intermediate state may be quite different from normal due to the preceding nuclear decay processes. The fact that internal electric field gradients of crystals must be employed, since no synthetic electric gradients of sufficient strength can be produced in the laboratory, makes a precise measurement of the electric quadrupole moment incomparably more difficult than the determination of the nuclear magnetic moment.

The static quadrupole attenuation coefficients for the time-integrated correlation (eqns. (11) and (12)) are again obtained from eqns. (27) to (31) by integration.

Since

$$I(\omega_e\tau_N)_{\tau_0} \equiv \frac{\int_0^{\tau_0} \exp(-t/\tau_N) \cdot \cos\omega_e t \cdot dt}{\int_0^{\tau_0} \exp(-t/\tau_N) \cdot dt}$$

$$= \frac{1}{1+(\omega_e\tau_N)^2} \frac{1-\exp(-\tau_0/\tau_N) \cdot \cos\omega_e\tau_0}{1-\exp(-\tau_0/\tau_N)} \quad . \quad . \quad . \quad (32)$$

(terms multiplied with $\omega_e\tau_N$ disappear, because $+\omega_e$ and $-\omega_e$ are equally probable), which becomes

$$I(\omega_e\tau_N)_\infty \equiv \frac{1}{1+(\omega_e\tau_N)^2} \quad \text{for} \quad \tau_0 \gg \tau_N, \quad . \quad . \quad . \quad (33)$$

the integral attenuation coefficients $G_{2k}(\omega_e{}^0\tau_N)_{\tau_0}$ and $G_{2k}(\omega_e{}^0\tau_N)_\infty$ can be written down immediately by replacing in eqns. (27) to (31) all the $\cos\omega_e t$ by $I(\omega_e\tau_N)_{\tau_0}$ and by $I(\omega_e\tau_N)_\infty$, respectively. To illustrate the behaviour of $G_{2k}(\omega_e{}^0\tau_N)_\infty$ with increasing quadrupole interaction $\omega_e{}^0\tau_N$, the coefficients $G_2(\omega_e{}^0\tau_N)_\infty$ and $G_4(\omega_e{}^0\tau_N)_\infty$ are plotted in fig. 7 for $I_B=2$.

The limiting values $G_{2k}(\infty)_\infty$ of the attenuation coefficients (table 2) for the integral correlation and for axially symmetric electric fields can easily be calculated from the eqns. (27) to (31). Rhombic fields ($\epsilon\neq0$) may give somewhat smaller values for $G_{2k}(\infty)_\infty$ (see table 2).

Table 2. Limiting Values of the Integral Attenuation Coefficients $G_{2k}(\infty)_\infty$ for Static Quadrupole Interaction in Isotropic Sources[46] and Minimum Values $G_{2k}(\min)$ for Static Interactions

I_B	$G_2(\infty)_\infty$	$G_4(\infty)_\infty$	$G_2(\infty)_\infty$	$G_4(\infty)_\infty$	$G_2(\min.)$	$G_4(\min.)$
	Axially symmetric fields		Rhombic fields : $\epsilon\neq0$			
1	0·60	—	0·40	—	0·20	0·11
$\frac{3}{2}$	0·20	—	0·20	—	0·20	0·11
2	0·37	0·46	0·29	0·29	0·20	0·11
$\frac{5}{2}$	0·20	0·11	$0·20+0·24\epsilon^2$		0·20	0·11
3	0·31	0·27	$\sim0·258$ for $\epsilon=1$		0·20	0·11

4.4.2. *Single Crystal Sources (Anisotropic Sources)*

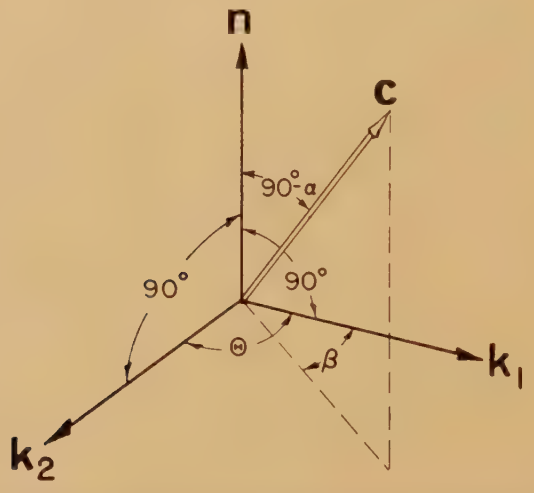
Instead of changing the effectiveness of the quadrupole interaction in a polycrystalline source by variation of the time delay, one can use single crystal sources to do the same by changing the relative orientation of the crystallographic axis with respect to the propagation directions of the nuclear radiations[55,56]. For simplicity we restrict ourselves to

axially symmetric crystalline gradient fields. The angular correlation function is again of the general form (15), since the existence of a crystallographic symmetry axis introduces a preferred direction at the source. Again it follows from the general considerations of § 4.1 that the unperturbed angular correlation is observed if the symmetry axis of the crystal is parallel to the propagation direction of one of the nuclear radiations (' parallel field ' case). Also, for symmetry reasons, the correlation function is given by a relatively simple expression, if the crystal axis is perpendicular to $\mathbf{k}_1$ and $\mathbf{k}_2$[46, 56],

$$W(\Theta) = \sum_{l=0}^{l_{\max}} B_{2l} \cdot \exp(i2l\Theta), \quad \ldots\ldots \quad (34)$$

where the B_{2l} are coefficients depending upon the radiation *and* the interaction parameters. In general, the B_{2l}, which are real in this particular case, cannot be written as a product of an attenuation factor and the coefficient b_{2l} of the unperturbed correlation.

Fig. 11



The position of the symmetry axis **c** with respect to the propagation directions $\mathbf{k}_1$ and $\mathbf{k}_2$.

To discuss the influence of the relative orientation of the crystal axis (axially symmetric field) on the angular correlation, we describe the position of the crystal axis **c** with respect to $\mathbf{k}_1$ and $\mathbf{k}_2$ by two angles α and β (see fig. 11). The correlation function (15) now becomes a function of α and β. To simplify the discussion we just consider the dependence of the anisotropy $A(\alpha, \beta)$ on the relative orientation of **c**. Numerical calculations of $A(\alpha, \beta)$ have been carried out[56] for gamma–gamma cascades observed with equally sensitive detectors and which involve transitions of multipole order not higher than quadrupole radiation.

The anisotropy is then

$$A(\alpha,\beta)=\frac{W(\alpha,\beta\,;\ \Theta=180°)}{W(\alpha,\beta\,;\ \Theta=90°)}-1, \quad . \quad . \quad . \quad . \quad (35\,a)$$

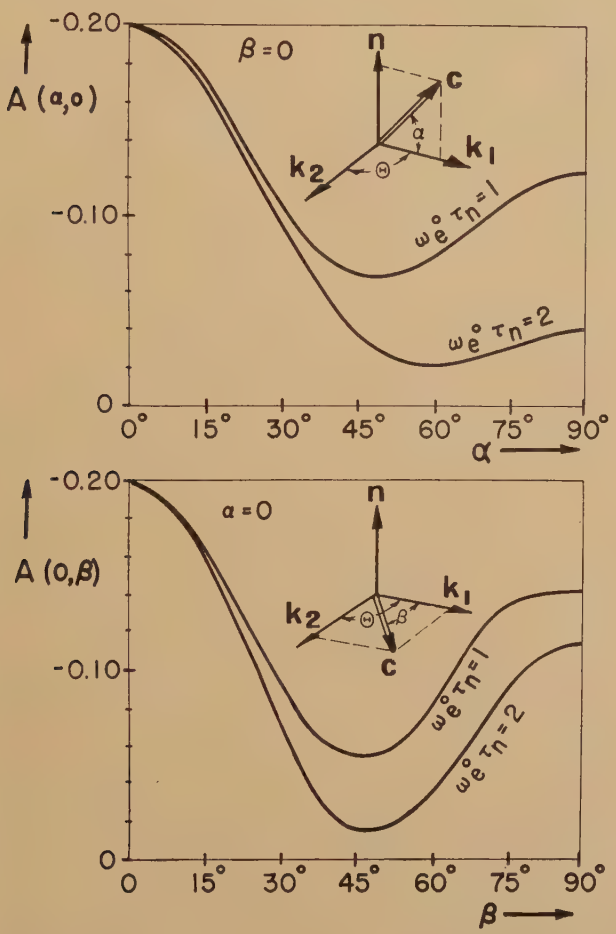
where

$$W(\alpha,\beta\,;\ \Theta=180°)=\sum_{n=0}^{4} r_{2n}\,.\cos^{2n}\alpha\,.\cos^{2n}\beta, \quad . \quad . \quad (35\,b)$$

$$W(\alpha,\beta\,;\ \Theta=90°)=s_0+s_2\cos^2\alpha+s_4\cos^4\alpha+s_8\cos^4\alpha\cos^2\beta\sin^2\beta$$
$$+s_{10}\cos^6\alpha\cos^2\beta\sin^2\beta+s_{16}\cos^8\alpha\,.\cos^4\beta\sin^4\beta. \quad (35\,c)$$

The coefficients r_{2n} and s_{2n} depend upon the radiation characteristics and $\omega_e{}^0\tau_N$. They are tabulated in the extensive theoretical work by

Fig. 12



Anisotropy of an angular correlation (e.g. ^{111}Cd gamma cascade) with static quadrupole interaction as function of the relative orientation of the symmetry axis **c** of the electrostatic gradient (according to Alder *et al.* reference (56)).

Alder *et al.*[56]. As an example, the anisotropy of a particular gamma–gamma cascade (^{111}Cd) as a function of the relative orientation of **c** is plotted in fig. 12 for different strengths of the quadrupole interaction $\omega_e{}^0\tau_N$.

Again a measurement of the anisotropy for various orientations of a single crystal source allows one to determine $\omega_e{}^0$ and to extract a value for the electric quadrupole moment of the intermediate nuclear state involved. Up to the present such a measurement has been accomplished in the case of ^{111}Cd (see Part II).

4.5. *Combined Static Magnetic and Electric Interaction*

One difficulty in measuring the quadrupole interaction always remains and is due to the fact that the strength of the electric field gradient in crystals cannot be varied. One has to change other parameters, such as the orientation of the crystal axis or the delay time, to obtain information on this type of interaction. A different approach, eliminating the necessity of growing single crystals or of using delayed coincidence techniques, has been suggested[56] and makes use of a combined electric and magnetic interaction. By measuring the anisotropy of a polycrystalline source with non-vanishing electric quadrupole interaction in a magnetic field, perpendicular to $\mathbf{k}_1$ and $\mathbf{k}_2$, the magnetic Larmor precession ω_m and the quadrupole interaction $\omega_e{}^0$ can be determined in one experiment. Unfortunately this case has not yet been treated theoretically, except for $\omega_m \gg \omega_e{}^0$ and $\omega_m \ll \omega_e{}^0$[46].

Another possible arrangement makes use of a suitable single crystal with its symmetry axis parallel to a variable magnetic field $\mathbf{H}^0$. The theory of this case of a parallel magnetic and electric field has been worked out by Alder *et al.*[56]. Again the angular correlation depends upon the relative orientation of the common field axis with respect to $\mathbf{k}_1$, $\mathbf{k}_2$ and also upon the magnetic and electric interaction parameters. For the case that the two field axes are normal to the plane ($\mathbf{k}_1$, $\mathbf{k}_2$), the angular correlation assumes the simple form (34). To illustrate the behaviour of the angular correlation as a function of the two interaction parameters ω_m and $\omega_e{}^0$, the coefficient B_2 of the particular angular correlation function $W(\Theta)=1+B_2\cos 2\Theta$ is plotted against the Larmor frequency ω_m (proportional to H_0) for different values of $\omega_e{}^0\tau_N$ as parameter (fig. 13).

It is interesting to note that for a particular value of $\omega_m\tau_N$ (or of the external magnetic field H_0) the angular correlation is partially restored. As pointed out by Alder and collaborators[56], this behaviour can be understood qualitatively by a semi-classical picture. As mentioned before, the electric quadrupole interaction causes the nucleus to precess in both directions around the symmetry axis with equal probability, whereas the magnetic Larmor precession is unidirectional. In the case of the combined electric and magnetic interaction the two precessions are superimposed and if the magnetic field is of such a

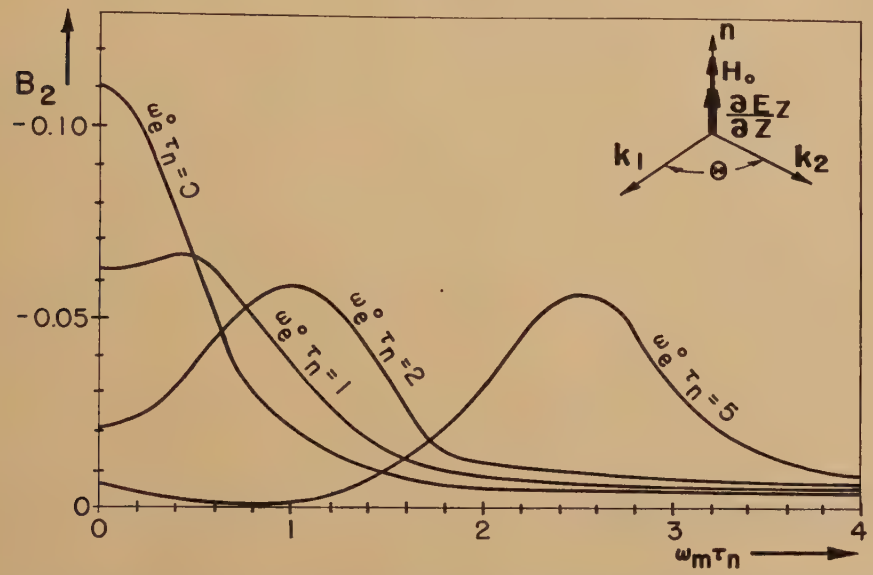
strength that $\omega_m = \frac{1}{2}\omega_e{}^0$ a maximum cancellation of the effects of the precession results, which gives rise to a maximum of the anisotropy.

4.6. *Time-dependent Interactions*

4.6.1. *Time-dependent Quadrupole Interaction in Liquids*

The environment of a given nucleus in a liquid is in a state of continual change, in a random manner, due to the Brownian motion. Thus, if the local configuration of the constituents of the liquid gives rise to an interaction with the nucleus, the axis of the interacting field changes continually and randomly in direction. However, for a very short time

Fig. 13



The coefficient B_2 for combined magnetic and electric interaction, the magnetic field and the symmetry axis of the electrostatic gradient both being perpendicular to the plane $(\mathbf{k}_1, \mathbf{k}_2)$ (Alder *et al.*, reference (56)).

interval, characterized by the correlation time τ_c, the local configuration of a given nucleus can be considered as constant, and not very dissimilar to the crystalline environment of a nucleus in a solid. The correlation time τ_c, which is of the order of 10^{-11} sec in water, is intimately related to the characteristic time of Debye's theory of dielectric dispersion in polar liquids[57]. If the nuclear lifetime τ_N is much larger than τ_c the interaction axis is in many uncorrelated directions during the nuclear lifetime, and thus no preferred direction exists at the site of the nucleus. Randomly fluctuating interacting fields can provide a rather powerful mechanism for inducing transitions between the different m-sublevels defined with respect to the propagation direction of the first nuclear radiation. For long enough nuclear lifetimes τ_N this 'relaxation' mechanism may lead to complete destruction of the angular correlation, which in general cannot be restored by a magnetic decoupling field.

The effect of time-dependent fields on nuclear angular correlations has been treated theoretically by Abragam and Pound[46]. For the time-dependent quadrupole interaction the coefficients G_{2k}, which are characteristic of the attenuation of the angular correlation (which, of course, is of the form (13)), are

$$G_{2k}(t)=\exp(-\lambda_{2k}t), \qquad (36)$$

where

$$\lambda_{2k}=\frac{3}{80}\left(\frac{eQ}{\hbar}\right)^2\left\langle\left(\frac{\partial E_z}{\partial z}\right)^2\right\rangle_{\mathrm{Av}}\cdot\tau_c\frac{2k(2k+1)[4I_B(I_B+1)-2k(2k+1)-1]}{I_B^2(2I_B-1)^2} \qquad (37)$$

$(eQ/h)^2\langle(\partial E_z/\partial z)^2\rangle_{\mathrm{Av}}$ is the mean square value of the quadrupole interaction. The time integrated value of $G_{2k}(\lambda\tau_N)_{\tau_0}$, applicable for a resolving time τ_0 without time delay, is

$$\left.\begin{aligned} G_{2k}(\lambda_{2k}\tau_N)_{\tau_0}&=\frac{1}{1+\lambda_{2k}\tau_N}\,\frac{1-\exp(-\tau_0/\tau_N)\exp(-\lambda_{2k}\tau_0)}{1-\exp(-\tau_0/\tau_N)} \\ \text{and}\quad G_{2k}(\lambda_{2k}\tau_N)_{\infty}&=\frac{1}{1+\lambda_{2k}\tau_N}\qquad\text{for}\qquad \tau_0\gg\tau_N. \end{aligned}\right\} \qquad (38)$$

For a liquid source, G_{2k} then becomes a function of the correlation time τ_c which is roughly proportional to the macroscopic viscosity η of the liquid. A similar formula holds if there is a time-dependent magnetic interaction in liquids, which, however, causes in practically all cases a negligibly small attenuating effect.

4.6.2. *Electronic Paramagnetic Relaxation*[46]

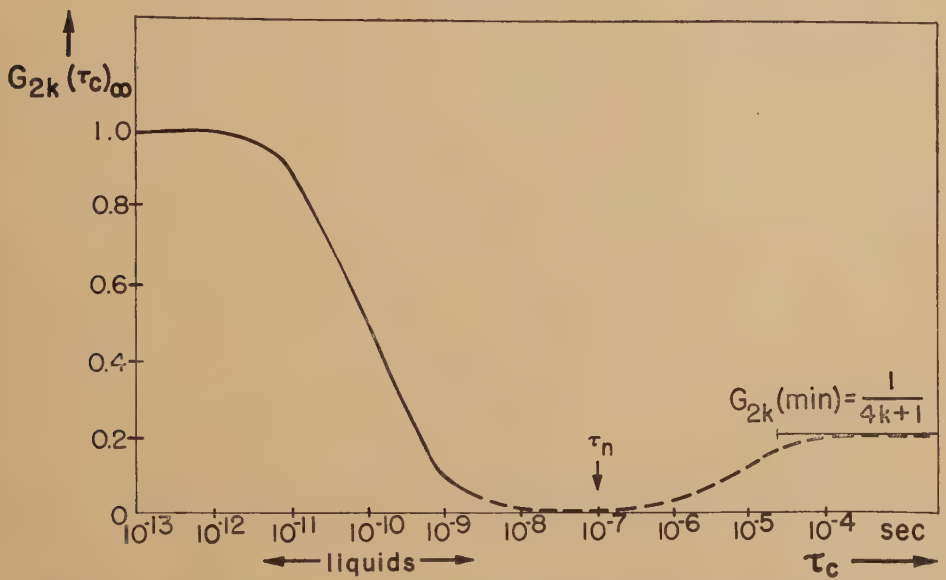
The influence of the magnetic hyperfine coupling on the correlation process was considered before with the important assumption that the electronic states giving rise to the I–J coupling were entirely stationary or, in other words, that J is a constant of the motion, at least over a time interval comparable to τ_N. In general, however, the atom or ion is a constituent of a solid and a coupling of the electronic states with the lattice must exist. In particular, the thermal vibrations of the lattice give rise to a time-dependent coupling with the electronic states of the ions. For paramagnetic ions, whose magnetic behaviour is entirely due to the electron spins, this electronic-state lattice interaction is caused by the spin–orbit coupling. The result of this spin–lattice interaction is a continual random change of the orientation of the electronic spin angular momentum **S**, equivalent in effectiveness on the angular correlation to a continually re-orienting magnetic field. Again, Abragam and Pound[46] showed how this interaction may be treated theoretically. The influence on the angular correlation can be expressed in terms of the attenuation coefficients $G_{2k}(t)=\exp(-\lambda'_{2k}t)$ where

$$\lambda'_{2k}=\tfrac{2}{3}\tau_s\omega_s^2 I_B(I_B+1)S(S+1)[1-(2I_B+1)W(I_B 1 2k I_B/I_B I_B)]. \qquad (39)$$

ω_s is a measure of the interaction energy $\hbar\omega_s\mathbf{I}\cdot\mathbf{S}$ between the nucleus and the magnetic shell moment due to the electron spin, and τ_s is the relaxation time of the electron spin. W is a so-called Racah coefficient[58],

a number which depends upon I_B and the index $2k$. Tables of these numbers were recently computed[59]. Expression (39) is valid only if $\omega_s \tau_s \ll 1$. τ_s is a function of the temperature of the lattice. In general, it decreases with increasing temperature. Hence, if this electronic paramagnetic relaxation is responsible for an attenuated angular correlation, the anisotropy should be temperature dependent. For $\tau_s < 5 \times 10^{-12}$ sec a magnetic decoupling field of about 10 000 gauss should partially restore the angular correlation affected by the spin relaxation in contradistinction to the quadrupole relaxation mechanism in liquids.

Fig. 14



Qualitative behaviour of the attenuation coefficients $G_{2k}(\tau_c)$ as a function of the correlation time (Abragam and Pound, reference (46)).

4.6.3. *The Transition from the Time-dependent to the Static Interaction Mechanism*

All the expressions for the time-dependent attenuation mechanisms were derived with the condition $t \gg \tau_c$, which means practically $\tau_N \gg \tau_c$ and $\tau_0 \gg \tau_c$. For a correlation time τ_c much larger than the lifetime τ_N of the intermediate nuclear state the attenuation mechanism can no longer be considered as time-dependent. In fact the environment of the nucleus will then be stationary during the nuclear lifetime and the resulting interaction is a static one, for which a minimum correlation exists. The qualitative behaviour[46] of the attenuation coefficients G_{2k} with increasing correlation time τ_c is illustrated in fig. 14. In the region where τ_c is of the order of magnitude of τ_N, that is where the interaction is described appropriately neither by time-dependent nor by static interaction mechanisms, the validity of the curve is obviously limited.

Part II. Experimental

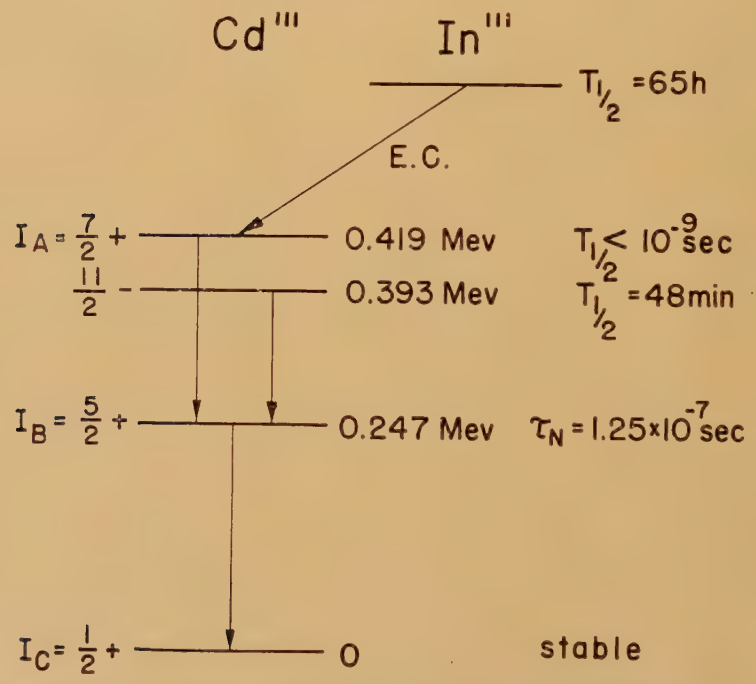
§ 5. The Angular Correlation of the ^{111}Cd Gamma Rays

5.1. *Effects of the Chemical and Physical State of the Radioactive Source*

5.1.1. *The ^{111}Cd Gamma–Gamma Cascade following the Electron Capture Decay of ^{111}In*

The first experimental evidence of an influence of extranuclear parameters was found by investigating the angular correlation of the ^{111}Cd gamma rays which follow the electron capture decay of ^{111}In (fig. 15). The intermediate nuclear level involved in this gamma cascade has a lifetime of $\tau_N = 1{\cdot}25 \times 10^{-7}$ sec[60,61], long enough that one expects an appreciable attenuation of the angular correlation by interaction of the nuclear moments with extranuclear fields.

Fig. 15



The decay of ^{111}In to ^{111}Cd and the decay of the ^{111m}Cd isomer.

The first attempts to show that the ^{111}In–^{111}Cd angular correlation, as measured for instance with polycrystalline $InCl_3$ sources, is influenced by interactions of the nucleus with its environment were made by Roberts and Steffen[62]. It was expected that this gamma–gamma correlation would be appreciably more attenuated when observed with

a delay ($t=1{\cdot}3\times10^{-7}$ sec, $\tau_0=1{\cdot}2\times10^{-7}$ sec) than when observed without delay. The difference actually found was so small that the effect was considered not to be real.

The possibility of the influence of extranuclear fields on angular correlation experiments had been discussed previously by Goertzel[49], Brady and Deutsch[17], and by Sunyar *et al.*[63] and experiments to observe the influencing effects by applying external magnetic fields and by using sources of different chemical compositions had been reported by Brady and Deutsch[17] in their pioneer work on angular correlation. All these experiments were based on the assumption that a modification of the environment (chemical compounds of different structure) resulting in different static fields at the nucleus or a variation of the delay time would cause different degrees of attenuation of the angular correlation.

5.1.2. *The Role of the Recovery of the Excited or Ionized* ^{111}Cd *Atom*

The first successful experiments which definitely established an influence of the extranuclear environment on angular correlation measurements[64] were done in Zürich, Switzerland, and were instigated by an idea first discussed by Frauenfelder[65], who noticed that the ground state of the atomic shell of many nuclei which exhibit an angular correlation is a $^1S_0$ or $^3P_0$ state. Both electronic states have zero magnetic moment ($J=0$) and hence there is no magnetic interaction between shell and nucleus. However, if the atom is in a state other than its non-magnetic ($J=0$) ground state for a time comparable with the intermediate nuclear lifetime, e.g. if it is excited or ionized, a perturbation of the angular correlation due to the magnetic I–J coupling is expected. As a matter of fact, the atomic shells of nuclei whose radiations exhibit the angular correlation will usually be in excited or ionized states as a result of preceding nuclear decay processes. Interactions of the radiation emitted by the nucleus with the electron shell (e.g. α-decay, β-decay), direct interaction of the shell electrons with radioactive nuclei (e.g. electron capture, internal conversion) or transfer of momentum (nuclear recoil) may lead to greatly disturbed atomic states. In general, the recovery is fast and the atom regains its ground state within a short recovery time T_r by emitting the excess energy in form of photons (light quanta, x-rays), phonons (radiationless transfer of energy to the lattice), by emission of electrons from the shell (e.g. Auger-effect), or by capturing an electron (de-ionization). While the atomic recovery process goes on, a strong interaction between the nuclear moments and the electron shell is to be expected, and if there is an appreciable probability for the nucleus to remain in its intermediate state B during this time, the angular correlation is perturbed considerably. For this to happen two more conditions must be fulfilled: (i) the intermediate nuclear level B must be reached in a time much shorter than the atomic recovery time T_r and (ii) the interaction of the (in general non-stationary) electron shell

with the nucleus in its intermediate state must be large enough to cause an appreciable re-orientation of I_B during the time T_r where the atomic recovery goes on as well as during the nuclear lifetime τ_N. If we describe the nucleus-excited shell interaction by some characteristic frequency ω, the condition required to observe attenuation effects on angular correlations is (*a*) $T_r\omega > 0{\cdot}1$ *and* (*b*) $\tau_N\omega > 0{\cdot}1$ (see § 4.1).

For a free atom T_r depends only upon the electronic configuration of the shell and the initial excitation or ionization caused by the nuclear decay process. In contradistinction, if the disturbed atom is in a solid or a liquid the interaction with its environment, e.g. the neighbouring atoms in a crystal lattice, the free electrons in a metal, the randomly moving neighbours in a liquid, will affect the recovery time T_r in a manner which will be quite different for different environments. Qualitatively one would expect the recovery time to be short if the excited atom finds itself in a metal, since the hole (resulting e.g. from electron-capture or beta-decay) can move in the full band. Crude estimates suggest a recovery time of less than about 10^{-12} sec in this case : and condition (*a*) is normally not satisfied. Hence a metallic source should display an angular correlation which is not affected much as a result of the de-excitation processes of the shell following nuclear decay. Nevertheless other perturbing fields may still be present, once the atom has reached its ground state (e.g. due to the crystalline structure of the metal). The situation is quite different if the excited atom is in an insulator, particularly in an ionic crystal, where it can act as an impurity centre, which, by virtue of the great radial extension of the excited electron orbits, has a rather long decay time, in general[66,67]. In such a case T_r may be of the order of microseconds and condition (*a*) may well be fulfilled. If condition (*b*) is also satisfied, an attenuated angular correlation is the result.

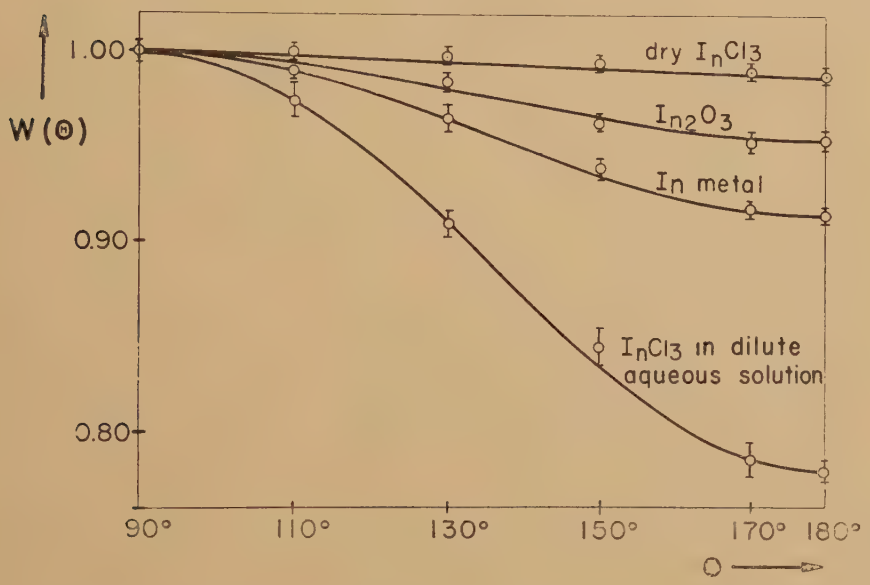
The Swiss workers found indeed a large difference of the angular correlation of the ^{111}Cd gamma rays depending upon whether they used ' metallic ' sources or ' ionic crystal ' sources. The metallic sources were prepared by evaporating simultaneously the radioactive ^{111}In and metallic Ag onto a heated surface, thus imbedding the ^{111}In in the Ag lattice. The maximum angular correlation measured with such an Ag source[68] was well represented by the correlation function

$$W(\Theta) = 1 - (0{\cdot}142 \pm 0{\cdot}004) P_2(\cos\Theta).$$

The corresponding anisotropy of $A = -0{\cdot}200 \pm 0{\cdot}006$ was much larger than any values previously found[62,69]. On the other hand, sources where the ^{111}In was imbedded in ionic crystals showed a very small anisotropy if any : $A = -0{\cdot}02 \pm 0{\cdot}02$[64]. Essentially identical results were obtained with sources prepared by electrolytic deposition and crystallization from solutions[70–72]. Figure 16 shows the pronounced influence of the chemical and physical structure of the ^{111}In source on the angular correlation of the ^{111}Cd gamma rays.

In table 3 are summarized the anisotropy values of the ^{111}In–^{111}Cd cascade as measured with solid ^{111}In sources of different structure and chemical composition, together with the attenuation coefficients G_2 extracted from the experimental data. To calculate these coefficients the true, unperturbed correlation must be known. There is good evidence (see § 5.1.4) that the unperturbed ^{111}Cd angular correlation is given by $W(\Theta)=1-(0{\cdot}175\pm0{\cdot}010)P_2(\cos\Theta)$ with a corresponding anisotropy of $A=-0{\cdot}245\pm0{\cdot}015$. Consequently this value has been used as the basis for the calculations of the experimental G_2 values.

Fig. 16



The influence of the chemical and physical structure of the ^{111}In source on the angular correlation of the ^{111}Cd gamma rays (R. M. Steffen, references (71, 72)).

The experimental results are essentially consistent with the aforementioned considerations relating the different attenuation of the angular correlation to the difference of the atomic recovery time in metals and in ionic crystals. However, only a part of the experimental results on solid sources can be explained in this way. Two facts require further consideration.

(i) The large anisotropy values ($A=-0{\cdot}200$) are found only with 'thick' Ag sources. Thin ($<10^4$ Å) Ag films display less anisotropic correlations, which depend upon the thickness of the metal film. Also, sources where the ^{111}In was imbedded in other metals (e.g. Cu, Au, Cd, In) exhibit considerably smaller anisotropy values ranging from

Table 3. Influence of Extranuclear Fields on the Angular Correlation of the ^{111}Cd Gamma Rays. I: Anisotropy values observed with solid ^{111}In sources of different chemical composition and structure

Form of the radioactive ^{111}In source	Anisotropy	G_2	Ref.
Metallic sources:			
^{111}In in Ag (double-stream evaporation), thickness of Ag foil $>10^4$ Å	$-0{\cdot}200 \pm 0{\cdot}006$	$0{\cdot}82 \pm 0{\cdot}04$	a
^{111}In in Ag (double-stream evaporation), thickness of Ag foil $\lesssim 10^4$ Å	$-0{\cdot}170 \pm 0{\cdot}014$	$0{\cdot}69 \pm 0{\cdot}10$	b
^{111}In in Ag (double-stream evaporation), thickness of Ag foil $\lesssim 10^4$ Å	$-0{\cdot}106 \pm 0{\cdot}019$	$0{\cdot}42 \pm 0{\cdot}09$	b
^{111}In in Ag (double-stream evaporation), thickness of Ag foil $\lesssim 10^2$ Å	$-0{\cdot}050 \pm 0{\cdot}021$	$0{\cdot}19 \pm 0{\cdot}07$	b
^{111}In in Ag (electrodeposited)	$-0{\cdot}18 \pm 0{\cdot}02$	$0{\cdot}73 \pm 0{\cdot}10$	c
^{111}In in Ag (^{111}In formed in Ag lattice by ^{109}Ag (α, 2n) ^{111}In reaction)	$-0{\cdot}18 \pm 0{\cdot}01$	$0{\cdot}73 \pm 0{\cdot}0$	d
^{111}In in Ag, same source as above, heat treated at 500°c for 6 h.	$-0{\cdot}05 \pm 0{\cdot}02$	$0{\cdot}20 \pm 0{\cdot}07$	d
^{111}In in In metal powder	$-0{\cdot}095 \pm 0{\cdot}005$	$0{\cdot}37 \pm 0{\cdot}03$	e
^{111}In in In metal (electrodeposited)	$-0{\cdot}056 \pm 0{\cdot}007$	$0{\cdot}21 \pm 0{\cdot}06$	f
^{111}In In metal, polycrystalline, isotropic source	$-0{\cdot}085 \pm 0{\cdot}003$	$0{\cdot}33 \pm 0{\cdot}02$	d
^{111}In in Cd (double-stream evaporation)	$-0{\cdot}06 \pm 0{\cdot}02$	$0{\cdot}23 \pm 0{\cdot}08$	g
^{111}In in Sn (double-stream evaporation)	$-0{\cdot}06 \pm 0{\cdot}02$	$0{\cdot}23 \pm 0{\cdot}08$	g
^{111}In in Au (double-stream evaporation)	$-0{\cdot}06 \pm 0{\cdot}02$	$0{\cdot}23 \pm 0{\cdot}08$	g
^{111}In in Au (electrodeposited)	$-0{\cdot}072 \pm 0{\cdot}010$	$0{\cdot}28 \pm 0{\cdot}05$	d
^{111}In in Sb (double-stream evaporation)	$-0{\cdot}06 \pm 0{\cdot}02$	$0{\cdot}23 \pm 0{\cdot}08$	g
^{111}In in Fe (double-stream evaporation)	$-0{\cdot}06 \pm 0{\cdot}02$	$0{\cdot}23 \pm 0{\cdot}08$	g
^{111}In in Cu (electrodeposited)	$-0{\cdot}085 \pm 0{\cdot}010$	$0{\cdot}34 \pm 0{\cdot}05$	d
Non-metallic sources:			
^{111}In in LiF (double-stream evaporation)	$+0{\cdot}016 \pm 0{\cdot}026$	$0{\cdot}06 \pm 0{\cdot}10$	b
^{111}In in AgCl (double-stream evaporation)	$-0{\cdot}004 \pm 0{\cdot}01$	$0{\cdot}02 \pm 0{\cdot}05$	b, g
^{111}In in SiO (double-stream evaporation)	$+0{\cdot}003 \pm 0{\cdot}02$	$-0{\cdot}01 \pm 0{\cdot}10$	b
$InCl_3$, polycrystalline, dry (room temp.)	$-0{\cdot}012 \pm 0{\cdot}005$	$0{\cdot}04 \pm 0{\cdot}02$	f
$InCl_3$, polycrystalline, dry (540°c)	$-0{\cdot}022 \pm 0{\cdot}006$	$0{\cdot}08 \pm 0{\cdot}03$	f
InI_3, polycrystalline	$-0{\cdot}020 \pm 0{\cdot}006$	$0{\cdot}08 \pm 0{\cdot}03$	f
$In(C_3H_3OH)_3$, polycrystalline	$-0{\cdot}020 \pm 0{\cdot}006$	$0{\cdot}08 \pm 0{\cdot}03$	f
In_2O_3, polycrystalline	$-0{\cdot}045 \pm 0{\cdot}004$	$0{\cdot}18 \pm 0{\cdot}03$	f
$In(OH)_3$, gelatinous	$-0{\cdot}036 \pm 0{\cdot}007$	$0{\cdot}13 \pm 0{\cdot}04$	f
$In(OH)_3$, dry	$-0{\cdot}035 \pm 0{\cdot}006$	$0{\cdot}13 \pm 0{\cdot}03$	f
$In(NO_3)_3$, $3H_2O$, dry	$-0{\cdot}031 \pm 0{\cdot}010$	$0{\cdot}12 \pm 0{\cdot}05$	d
$In_2(SO_3)_3$, dry	$-0{\cdot}02 \pm 0{\cdot}02$	$0{\cdot}08 \pm 0{\cdot}08$	h

a. Aeppli, Frauenfelder, and Walter, 1951, *Helv. Phys. Acta*, **24,** 335.
b. Aeppli, Bishop, Frauenfelder, Walter, and Zunti, 1951, *Phys. Rev.*, **82,** 550.
c. R. M. Steffen, 1952, *Phys. Rev.*, **86,** 632.
d. R. M. Steffen, to be published.
e. Albers-Schönberg, Heer, Novey, and Ruetschi, 1953, *Phys. Rev.*, **91,** 199.
f. R. M. Steffen, 1953, *Phys. Rev.*, **90,** 1119.
g. H. Frauenfelder, 1952, *Annual Review of Nuclear Science*, Vol. 2.
h. J. C. Kluyver, and M. Deutsch, 1952, *Phys. Rev.*, **87,** 203.

$A=-0{\cdot}05$ to $A=-0{\cdot}09$. The corresponding attenuation coefficients, however, are all above the minimum value, $G_2(\min)=0{\cdot}20$, allowed for static interaction and isotropic sources.

(ii) The attenuation of the ^{111}Cd angular correlation in non-metallic sources in most cases is more pronounced ($G_2<G_2(\min)=0{\cdot}20$) than is theoretically possible with static interactions alone.

5.1.3. *The* ^{111}Cd *Angular Correlation and the Structure of Solid* ^{111}In *Sources*

The considerable attenuation found with most metal sources, except the ' thick ' Ag sources, can be explained satisfactorily in the following way[46,73]. It is still assumed (and all experimental data, so far, support this assumption) that, in metals, no attenuation effects are due to the recovery processes of the electronic shell. Nevertheless, inhomogeneous static electric fields (gradients) interacting with the electric quadrupole moment of the intermediate state can be responsible for a pronounced attenuation of the angular correlation. Such fields are certainly present in crystals of non-cubic symmetry like the tetragonal In or the hexagonal Cd. In the cubic crystal lattice of Cu or Au inhomogeneous static electric fields may also be present at the site of the ^{111}Cd nucleus formed by the K-capture decay of ^{111}In. The latter is imbedded as an impurity in these cubic lattices and the lattice will in general be distorted appreciably in the environment of the In–Cd ion, due to the presence of this foreign ion, unless it fits well into the host lattice. To a large degree the latter seems to be true in the case of the In–Cd imbedded in the cubic Ag lattice, where the attenuation of the angular correlation is small. Why there is such a difference between the Ag and Au sources, the two metals in general being considered quite similar, remains unclear.

The fact that the attenuation is more pronounced in the thin ($<10^4$ Å) Ag films is not too surprising since it is well known that thin metal films display strong anomalies in their electric and magnetic properties and may behave more like semiconductors rather than metals. Furthermore the crystalline structure of the thin Ag films is considerably distorted and lacks perfect cubic symmetry. In addition the ^{111}In impurity in the Ag lattice may diffuse to the grain boundaries of the microcrystals and there be subjected to strong electrostatic gradients. This assumption is corroborated by the results obtained with heat treated ' thick ' Ag films, in which the ^{111}In was produced by irradiation of the Ag foil ($2{\cdot}5\times10^4$ Å thick) with 20 MeV alpha particles. The ^{111}In is then formed inside the Ag lattice by an (α, 2n) reaction. In such nuclear reactions the atoms are irreversibly ejected from their normal positions in the lattice, leaving vacant lattice sites behind, and finally coming to rest in interstitial positions. Consequently the displaced ^{111}In atom may find itself in a position where large electric gradients are present. This should manifest itself in a strongly attenuated angular correlation.

Strangely enough, the anisotropy ($A=-0{\cdot}18\pm0{\cdot}01$) measured with such sources is rather close to the maximum value found with ' thick ' Ag sources. Evidently either the electric gradient at the site of the interstitial In impurity is unexpectedly small or, more probably, some ' healing ' of the ' radiation damage ' must have taken place. Such a ' healing ' process would involve a diffusion of the interstitial ^{111}In atoms in the Ag lattice until they find a vacant lattice site. This process should be enhanced in this particular case, since there are abnormally many vacant lattice sites in the neighbourhood of the displaced ^{111}In atom by virtue of the knock-on collisions of the α-particle which formed the ^{111}In atom and also by virtue of the motion of the recoiling ^{111}In atom itself. However, the ' healing ' process is not quite complete, which is shown by the fact that the anisotropy increases from $-0{\cdot}18\pm0{\cdot}01$ to $-0{\cdot}22\pm0{\cdot}01$ upon melting and recrystallization of the Ag source. An attempt to enhance the ' healing ' process by annealing the Ag foil for several hours at a temperature of 800°c failed. In fact the anisotropy displayed by the heat treated Ag foils was reduced to $-0{\cdot}06\pm0{\cdot}02$. Again, this effect could possibly be traced to the diffusion of the ^{111}In impurity atoms to the Ag grain boundaries during the high annealing temperature.

The consideration of the static quadrupole interaction as the main attenuation mechanism in the metal sources is also supported by the fact that none of the metal sources show an angular correlation with an anisotropy smaller than the ' hard core ' value for static quadrupole interaction (cf. table 2). Furthermore, magnetic decoupling experiments[55,72] on these metal sources failed to restore the angular correlation, indicating that the interaction in the metals must be electric. A direct proof for the static quadrupole interaction in In metal sources has been given by the Zürich group with their single crystal measurements[55], which will be discussed later.

The anisotropy values of the non-metallic solid sources (table 3), which correspond to attenuation coefficients considerably lower than the theoretically possible minimum value for static interaction, suggest the presence of a time-dependent interaction mechanism which provides for a complete disappearance of the angular correlation. Hence we are forced to conclude that the interaction of the nucleus with the atomic shell, which recovers relatively slowly in these non-metallic sources and which is in a non-stationary state during the lifetime of the intermediate nuclear state, must be described as a time-dependent interaction mechanism rather than a static one.

That the small anisotropy values of non-metallic ^{111}In sources are in fact due to the after-effects of the ^{111}In electron capture has recently been shown by Kraushaar and Pound[74] in an extremely interesting experiment. In order to have the ^{111}Cd nucleus in a well-defined environment, undisturbed by preceding nuclear decay processes, they measured the angular correlation of the two gamma rays emitted by the

48 min isomeric state of ^{111}Cd. The intermediate state of this gamma cascade is the same as the one involved in the gamma cascade following the electron capture of ^{111}In (see fig. 15). The results of these angular correlation measurements obtained with different ^{111}Cd sources are shown in table 4. As expected, the angular correlation is greatly reduced in some sources due to the static quadrupole interaction in the non-cubic crystal lattices, but none of the experimental attenuation coefficients G_2 are below the minimum value $G_2(\text{min})=0{\cdot}20$ for static interactions, an indication that the smaller values of G_2 found with the non-metallic ^{111}In sources are very probably due to the after-effects of the ^{111}In electron capture decay.

Table 4. Influence of Extranuclear Fields on the Angular Correlation of the ^{111}Cd Gamma Rays. II: Attenuation observed with different sources of the 48 min ^{111}Cd isomer (J. J. Kraushaar, and R. V. Pound, 1953, *Phys. Rev.*, **82,** 522).

Form of the radioactive ^{111}Cd source	G_2
Cd metal, liquid	$0{\cdot}97\pm0{\cdot}08$
$CdCl_2$, aqueous solution	$0{\cdot}88\pm0{\cdot}05$
CdO, polycrystalline, cubic lattice	$0{\cdot}63\pm0{\cdot}08$
Cd metal, solid, hexagonal lattice	$0{\cdot}40\pm0{\cdot}07$
$CdCl_2$, solid, hexagonal lattice	$0{\cdot}19\pm0{\cdot}06$

5.1.4. *The* ^{111}Cd *Angular Correlation with Liquid Sources*

Actually the static quadrupole interaction hypothesis was first invoked by M. Deutsch[75] to interpret measurements on aqueous solutions of $In_2(SO_4)_3$, which showed an only slightly perturbed correlation ($A=-0{\cdot}18\pm0{\cdot}02$), whereas dry polycrystalline sources of the same salt yielded a practically isotropic correlation ($A=0{\cdot}02\pm0{\cdot}02$). Extensive measurements[71, 72] on sources in the liquid state (see table 5) showed conclusively that the physical state (liquid) rather than the symmetry of the particular environment of the decaying nucleus in an aqueous solution is responsible for the smallness of the interaction, since all liquid sources of very different character displayed the large anisotropy. The role of the physical state of the radioactive source in the attenuation of angular correlation measurements is best demonstrated by figs. 17 and 18 where the anisotropy of InI_3 and In metal sources is plotted as a function of source temperature[71]. The sudden increase of the anisotropy at the melting point of the two source substances indicates that the change of state and not the temperature change is responsible for the larger anisotropy observed in the molten state. It is interesting to note that the metallic In source showed a smaller anisotropy before melting than after recrystallization. This difference was found[77] to

result from some preferred orientation of the microcrystals in the metallic In source, which was prepared by electrodeposition of In onto an In wire. After melting and recrystallization the source was perfectly isotropic as a whole. Thus the larger anisotropy ($A=-0{\cdot}085\pm0{\cdot}003$) corresponds to the case of the polycrystalline powder.

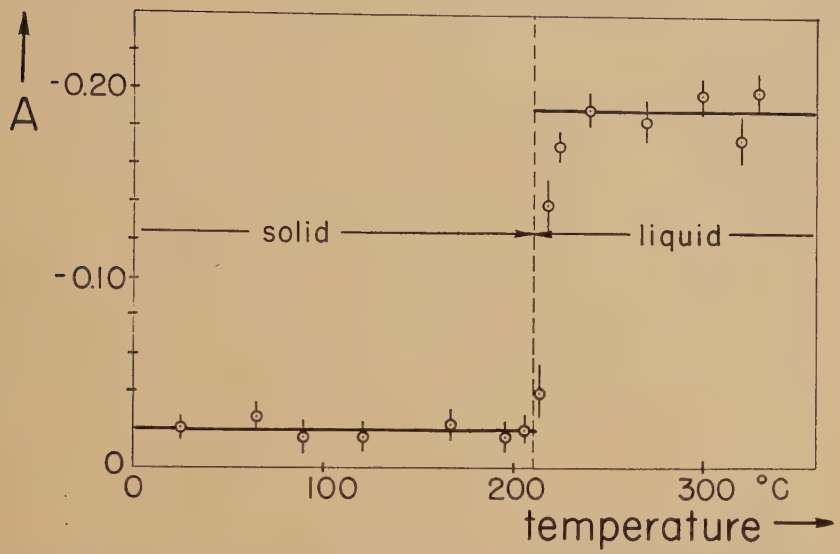
Table 5. Influence of Extranuclear Fields on the Angular Correlation of the ^{111}Cd Gamma Rays. III: Anisotropy values observed with different liquid in ^{111}In sources.

Form of the radioactive ^{111}In source	Anisotropy	G_2	Ref.
In metal, liquid (180°c)	$-0{\cdot}21\ \pm0{\cdot}005$	$0{\cdot}86\pm0{\cdot}05$	b, c, e
$In_2(SO_4)_3$, dilute aqueous solution	$-0{\cdot}18\ \pm0{\cdot}02$	$0{\cdot}73\pm0{\cdot}10$	d
$InCl_3$, dilute aqueous solution	$-0{\cdot}221\pm0{\cdot}005$	$0{\cdot}90\pm0{\cdot}02$	a, b
InI_3, dilute aqueous solution	$-0{\cdot}21\ \pm0{\cdot}01$	$0{\cdot}86\pm0{\cdot}05$	a
$In(NO_3)_3.3H_2O$, dilute aqueous solution	$-0{\cdot}21\ \pm0{\cdot}02$	$0{\cdot}86\pm0{\cdot}10$	a
$In(ClO_4)_3.8H_2O$, dissolved in ethyl alcohol	$-0{\cdot}21\ \pm0{\cdot}01$	$0{\cdot}86\pm0{\cdot}05$	a
InI_3, dissolved in xylene	$-0{\cdot}22\ \pm0{\cdot}01$	$0{\cdot}90\pm0{\cdot}05$	a
$In(NO_3)3H_2O$, dissolved in ethyl alcohol	$-0{\cdot}22\ \pm0{\cdot}01$	$0{\cdot}90\pm0{\cdot}05$	a
$In(C_9H_6ON)$, dissolved in chloroform	$-0{\cdot}20\ \pm0{\cdot}01$	$0{\cdot}82\pm0{\cdot}05$	a
InI_3, liquid salt (220°c)	$-0{\cdot}19\ \pm0{\cdot}02$	$0{\cdot}77\pm0{\cdot}10$	a, b
$InCl_3$, liquid salt (610°c)	$-0{\cdot}18\ \pm0{\cdot}03$	$0{\cdot}73\pm0{\cdot}14$	a
$InCl_3$, aqueous solution $+10^{21}$ Fe^{++} ions/cm^3 added	$-0{\cdot}215\pm0{\cdot}006$	$0{\cdot}88\pm0{\cdot}04$	a, b

a. R. M. Steffen, 1953, *Phys. Rev.*, **89,** 903.
b. R. M. Steffen, 1953, *Phys. Rev.*, **90,** 1119.
c. R. M. Steffen, unpublished data.
d. J. C. Kluyver, and M. Deutsch, 1953, *Phys. Rev.*, **87,** 203.
e. Albers-Schönberg, Heer, Novey, and Ruetschi, 1953, *Phys. Rev.*, **91,** 199.

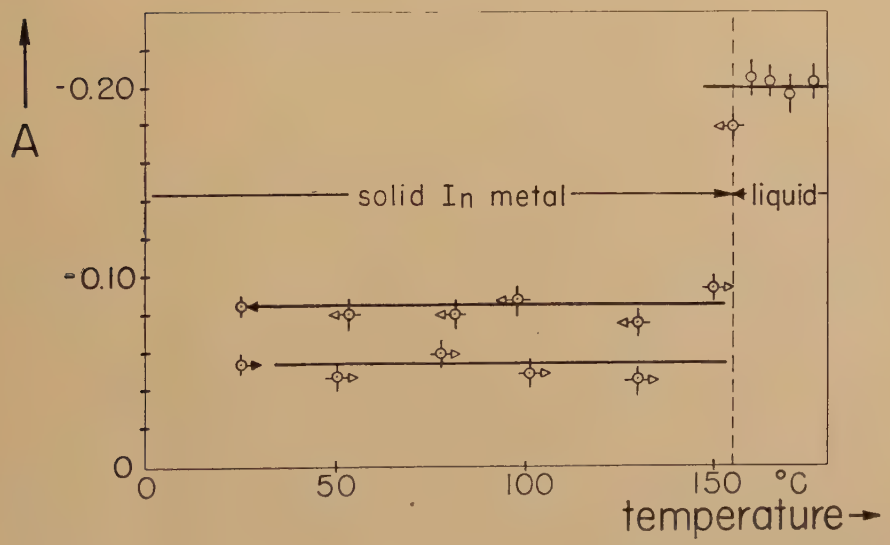
Two qualitative conclusions may be drawn from the large anisotropies measured throughout with sources in the liquid state: (*a*) In the continually and randomly fluctuating environment of an atom or an ion in a liquid the recovery of the Cd electronic shell from the preceding K-capture probably takes place in a time which is short compared to the nuclear lifetime (10^{-7} sec). (*b*) The difference in the electron configuration of the atoms in the solid and in the liquid state is insignificant for the cases considered. Consequently the large change of the anisotropy observed with the change of state of the radioactive source must be due to the difference of the local environment of a given atom in the crystalline structure of the solid and the randomly fluctuating local configuration in the liquid state. Hence the attenuation of the angular correlation as measured with most of the solid metal sources cannot be attributed to the fields due to the electronic configurations of the particular atom whose nucleus is involved in the gamma–gamma cascade. In general, nternal fields in a solid for which the crystalline structure, rather than

Fig. 17



The influence of the phase (solid–liquid) of dry InI_3 sources on the ^{111}Cd gamma–gamma correlation (R. M. Steffen, reference (71)).

Fig. 18



The influence of the phase (solid–liquid) of In metal sources on the ^{111}Cd gamma–gamma correlation (R. M. Steffen, reference (71)).

the electronic structure of the particular atoms is responsible, are electric rather than magnetic fields. In view of this fact the large increase of the anisotropy exhibited by the In metal sources after melting is another indication of the dominant role of the static quadrupole interaction in metallic sources of non-cubic structure.

In a non-viscous liquid, e.g. an aqueous solution or a molten metal, the correlation time τ_c is very short, of the order of 10^{-11} sec. and the attenuation of the angular correlation due to the randomly fluctuating electric gradients in the liquid (quadrupole relaxation mechanism) is expected to be small[45,46] (compare fig. 14) in agreement with the experimental results (table 5). However, for larger correlation times τ_c the relaxation mechanism in the liquid may become strong enough to influence the angular correlation substantially. The correlation time τ_c is roughly proportional to the macroscopic viscosity η of the liquid. As a crude approximation the following relationship between τ_c and η may be used[25,57]:

$$\tau_c = \frac{4\pi a^3}{3kT}\eta, \qquad \ldots\ldots\ldots \quad (40)$$

where a is the radius of the ions, atoms, or molecules in the liquid. Consequently the parameter λ_{2k} for the time-dependent quadrupole interaction, eqn. (42), and hence the degree of attenuation of the angular correlation, depends upon the macroscopic viscosity of the liquid source. Using the approximate expression (40) the attenuation coefficients $G_{2k}(\eta)\tau_0$ for the time-integrated angular correlation (38) as measured with viscous liquid sources become

$$G_{2k}(\eta)\tau_0 = \frac{1}{1+\lambda_{2k}(\eta)\tau_N}\,\frac{1-\exp\left[-\lambda_{2k}(\eta)\,.\,\tau_0\right]\exp\left(-\tau_0/\tau_N\right)}{1-\exp\left(-\tau_0/\tau_N\right)}, \qquad (41\,a)$$

$$\lambda_{2k}(\eta) = \frac{\pi}{20}\left(\frac{eQ}{\hbar}\right)^2\left\langle\left(\frac{\partial E_z}{\partial z}\right)^2\right\rangle_{\mathrm{Av}} \times \frac{2k(2k+1)[4I_B(I_B+1)-2k(2k+1)-1]}{{I_B}^2\,.\,(2I_B-1)^2}\,\frac{a^3}{kT}\,.\,\eta. \qquad (41\,b)$$

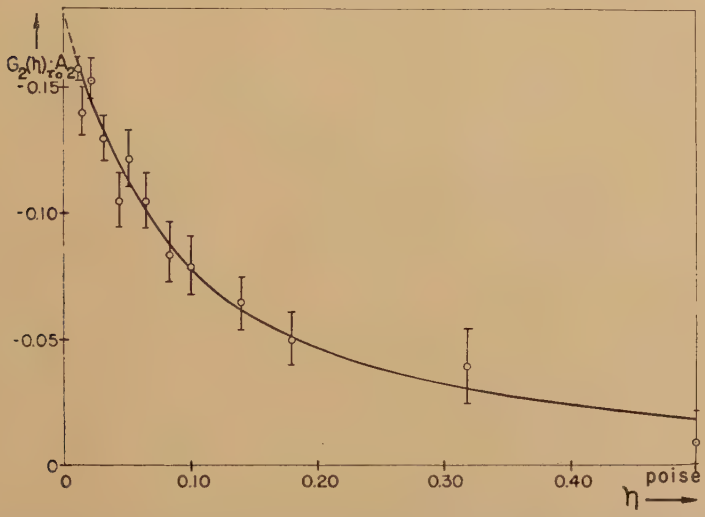
Experimental investigations of this time-dependent interaction mechanism in liquid ^{111}In sources of different viscosity were carried out by Hemmig and Steffen[79], and by the Zürich group[80]. The viscosity of the sources was varied by adding glycerine to very dilute aqueous $InCl_3$ solutions containing the radioactive ^{111}In, and the anisotropy $A(\eta)$ of the ^{111}Cd correlation was determined. In fig. 19 the coefficients

$$G_2(\eta)_{\tau_0}A_2 = \frac{A(\eta)}{1{\cdot}5-0{\cdot}5A(\eta)}$$

of the directional correlation function are plotted versus the macroscopic viscosity of the $InCl_3+H_2O+$glycerine source. The solid line in fig. 19 represents the theoretical expression (41), calculated with $A_2=-0{\cdot}18$, corresponding to an anisotropy of the unperturbed correlation of

$A=-0{\cdot}25$ and with a root mean square value of the quadrupole interaction of $eQ(\partial E_z/\partial z)/h \simeq 500$ Mc/s. The molecular radius a was assumed to be $1{\cdot}5\times10^{-8}$ cm. The anisotropy value of $-0{\cdot}25\pm0{\cdot}02$ extrapolated from the viscous source measurements corresponds closely to the angular correlation observed with In-metal single crystal sources ($A=-0{\cdot}23\pm0{\cdot}02$, see next section) in the parallel field orientation which is expected to yield the unperturbed correlation (cf. § 3.4). Furthermore, recent measurements with very short coincidence resolving times τ_0, such that $\tau_0\omega<0{\cdot}1$, yield the same large anisotropies ($A=-0{\cdot}27\pm0{\cdot}04$ and $A=-0{\cdot}24\pm0{\cdot}04$, cf. § 5.4). Thus there is satisfactory evidence to assume that the unperturbed angular correlation of the ^{111}In–^{111}Cd gamma rays is represented by $W(\Theta)=1-(0{\cdot}175\pm0{\cdot}010)\,.\,P_2(\cos\Theta)$ with an anisotropy of $A=-0{\cdot}245\pm0{\cdot}015$. Knowing the true angular correlation the attenuation factors arising from the extranuclear fields in the different ^{111}In sources can now be determined.

Fig. 19



The attenuation of the ^{111}Cd gamma–gamma correlation as a function of the macroscopic viscosity η of the liquid ^{111}In source ($InCl_3$ dissolved in a water–glycerine mixture) (Hemmig and Steffen, reference (79)).

Using the 48 min ^{111}Cd isomer as radioactive source Kraushaar and Pound[74] also measured the attenuation of this ^{111}Cd gamma–gamma correlation in a dilute aqueous solution of $CdCl_2$. Their result for the attenuation factor, $G_2=0{\cdot}88\pm0{\cdot}04$, is in excellent agreement with the correlation measurements using sources of ^{111}In in the form of dilute aqueous solutions of $InCl_3$, InI_3, etc. (cf. table 5).

The root mean square value of the quadrupole interaction extracted from the attenuation data of the ^{111}In–^{111}Cd angular correlation in

viscous sources must be considered a rather rough estimate; because (*a*) the approximation used for τ_c is a very crude one, and (*b*) the mean square value of the electrostatic gradient $\langle(\partial E_z/\partial z)^2\rangle_{\mathrm{Av}}$ is probably not the same for the somewhat different environments of the Cd ions in the various water–glycerine mixtures. The effect of fluctuating magnetic fields in the liquids considered here can probably be neglected. This assumption is strongly supported by the fact that the addition of a large concentration of paramagnetic ions (Fe^{+++}) to aqueous $InCl_3$ solutions did not cause any further attenuation of the angular correlation[72] (see table 5). The Zürich group[80] observed a similar viscosity dependence of the anisotropy by changing the temperature of a source of $InCl_3$ dissolved in glycerine.

The viscosity of a liquid radioactive source can be changed continuously and over a rather wide range without difficulty. Hence the measurement of the attenuation of angular correlations displayed by sources of different viscosities and an extrapolation to zero viscosity ($\tau_c \to 0$) should, in most cases, yield the unperturbed angular correlation. This procedure seems to offer a promising approach to the problem of finding the 'true' angular correlation of a nuclear cascade.

5.2. *The* ^{111}Cd *Gamma–Gamma Angular Correlation in an External Magnetic Field*

In the discussion of the experimental data on the ^{111}Cd gamma cascade it was recognized that the electric quadrupole interaction provides a consistent interpretation of the attenuation observed with sources of different chemical and physical structure. Yet, in many of these cases, the magnetic hyperfine coupling probably should not be neglected entirely. An indication of the presence of a weak magnetic coupling in a solid $In(OH)_3$ source was observed recently. By applying a magnetic decoupling field of 7500 gauss in the direction of one detector (parallel field case) a slight increase of the anisotropy from $-0{\cdot}035\pm0{\cdot}005$ to $-0{\cdot}050\pm0{\cdot}005$ was measured[81].

The investigation of the magnetic coupling is facilitated by the fact that external magnetic fields of strength sufficient to cause an appreciable attenuation of the ^{111}Cd cascade are easily produced. Fields of a few thousand gauss are adequate for cascades involving intermediate states of lifetimes of 10^{-7} sec or longer. From the attenuation and the azimuthal angular shift of the angular correlation, observed with a known magnetic field applied at right angles to the ($\mathbf{k}_1$, $\mathbf{k}_2$) plane, the magnetic moment μ of the intermediate nuclear state can be determined. Such a measurement, first proposed by Goertzel[49] and Brady and Deutsch[17], has been successfully performed on the ^{111}Cd cascade[82,78] and also very recently on the gamma–gamma cascade of the ^{204}Pb isomer[83] (cf. § 6).

To avoid ambiguities in the interpretation of the magnetic attenuation the sources used for such measurements should exhibit an angular

correlation completely unperturbed by crystalline and atomic fields in the source itself; or, if such an interaction is unavoidable, the interaction parameters must be well known. This requirement is not easy to satisfy in the case of ^{111}Cd. The Zürich group measured the magnetic moment of the 0·247 Mev level of ^{111}Cd with sources in which the radioactive parent ^{111}In was imbedded in the cubic lattice of polycrystalline Ag foils[82]. The attenuation of the angular correlation of the ^{111}In–^{111}Cd cascade as a function of the strength of the (perpendicular) magnetic field is seen in fig. 20 (*a*), where the coefficient $B_2(H_0)=G^{(2)}(H_0)_{\tau_0} \cdot b_2$ of the angular correlation function $W(\Theta, H_0)=1+B_2(H_0)\cos 2\Theta$, which was measured with equally sensitive detectors for magnetic fields up to 8000 gauss, is plotted. Use can be made of eqn. (25) to extract from the experimental data the value of $\omega_m\tau_N$. From eqn. (7) and knowing the lifetime $\tau_N=(1{\cdot}25\pm0{\cdot}06)\times10^{-7}$ sec the value (but not the sign) of the magnetic moment μ_B of the intermediate level was computed: $\mu_B=-(0{\cdot}70\pm0{\cdot}12)\mu_0$ where μ_0 is the nuclear magneton $\mu_0=e\hbar/2$ Mc. The sign of μ_B was obtained by measuring the sign of the azimuthal shift $\Phi \simeq \omega_m\tau_N$ with detectors which were of unequal sensitivity for the two ^{111}Cd gamma rays.

Owing to the interaction of the nuclear quadrupole moment with the electrostatic gradients, which do not vanish completely in polycrystalline Ag sources (cf. table 3), the value of the magnetic moment obtained in this way must be corrected accordingly[46].

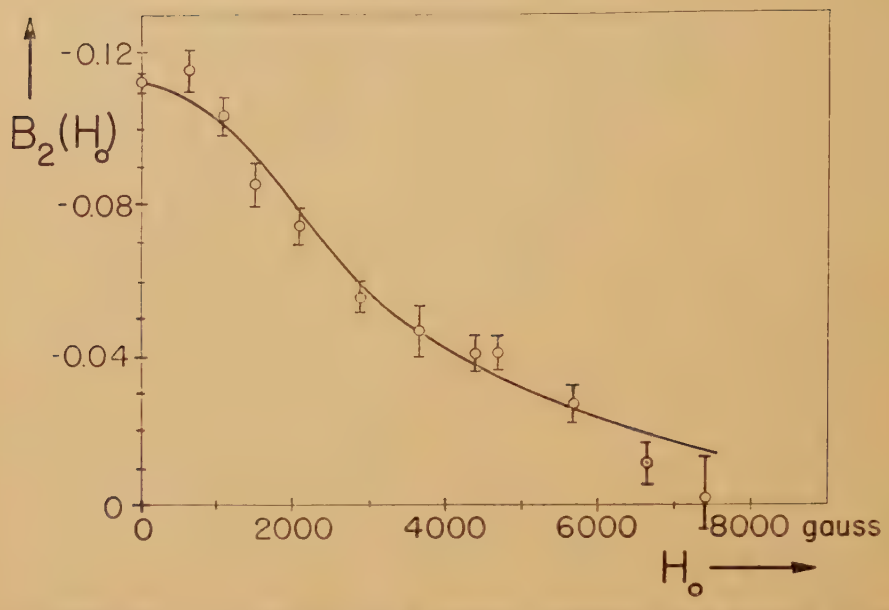
Steffen and Zobel[78], in their investigation of the magnetic moment of the excited ^{111}Cd state, made use of the relationship between μ, H_0 and the azimuthal shift of the correlation pattern about H_0. If H_0 is perpendicular to the propagation directions of the two gamma rays, and if other interactions can be neglected, the delayed angular correlation of the ^{111}Cd cascade is given by eqn. (23) or, in the Legendre polynomial expansion, by $W(\Theta, t)=1+A_2P_2[\cos(\Theta+\omega_m t)]$. In case the delayed correlation measurement is performed with a liquid source the time-dependent quadrupole interaction cannot be neglected:

$$W(\Theta, t)=1+A_2\exp(-\lambda 2t)\,P_2[\cos(\Theta+\omega_m t)].$$

In order to obtain the highest accuracy possible with present experimental techniques, the following method of measurement was used. The source (dilute aqueous solutions of $InCl_3$) was viewed by three NaI(Tl) scintillation detectors A, B and C, which are in the x–y plane with the source S (fig. 20 (*b*)). Coincidence analysers (resolving time $\tau_0=10^{-7}$ sec) registered coincidences between the detector pairs A–B and A–C with a constant delay T ($2{\cdot}50+10^{-7}$ sec or $3{\cdot}75\times10^{-7}$ sec) in the channel of detector A.

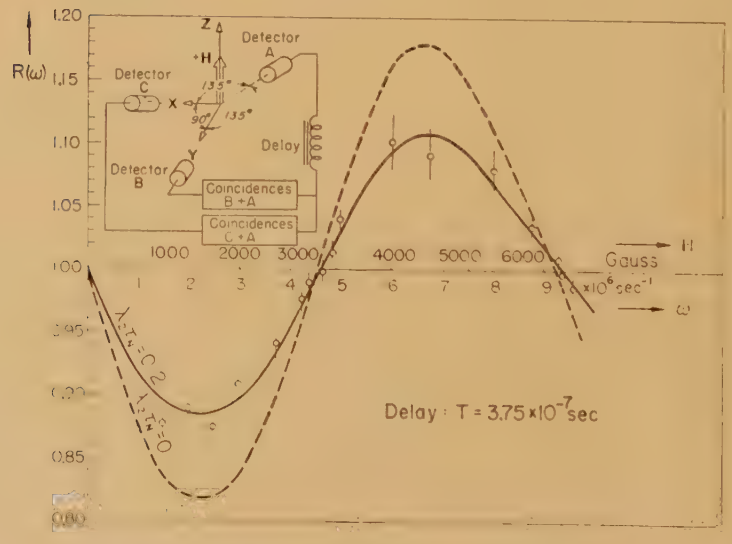
The (constant) angle subtended at the source by the axes of each detector pair was 135°. The ratio, $R=C_{135°}(+H)/C_{135°}(-H)$, of the coincidence counting rates, $C_{135°}(+H)$ and $C_{135°}(-H)$, was determined as a function of the magnitude of the magnetic field with its direction

Fig. 20 (*a*)



The coefficient B_2 of the ^{111}In–^{111}Cd angular correlation as a function of the strength of an external magnetic field applied perpendicularly to the plane $(\mathbf{k}_1, \mathbf{k}_2)$. The solid line is the theoretical curve, calculated for $\mu = -0{\cdot}70\,\mu_0$ (Aeppli *et al.*, reference (82)).

Fig. 20 (*b*)



The ratio $R(\omega_m) = W(3\pi/4, t, +\omega_m)/W(3\pi/4, t, -\omega_m)$ as a function of the Larmor frequency ω_m (theory) and as a function of the magnetic field strength H_0 (experimental points) (reference (78)).

upwards ($+H$) and downwards ($-H$), respectively. This ratio is given by

$$R(\omega_m)=\frac{W_{\tau_0}\left(\frac{3\pi}{4}, t, +\omega_m\right)}{W_{\tau_0}\left(\frac{3\pi}{4}, t, -\omega_m\right)}$$

$$=\frac{\int_{t-\tau_0}^{t+\tau_0}\left\{1+A_2\exp(-\lambda_2 t')P_2\left[\cos\left(\frac{3\pi}{4}+\omega_m t'\right)\right]\exp(-\lambda t')\right\}dt'}{\int_{t-\tau_0}^{t+\tau_0}\left\{1+A_2\exp(-\lambda_2 t')P_2\left[\cos\left(\frac{3\pi}{4}-\omega_m t'\right)\right]\exp(-\lambda t')\right\}dt'}.$$

The ratio $R(\omega_m)$, corrected for the finite resolution of the equipment is plotted in fig. 20 (*b*) for $\lambda_2\tau_N=0{\cdot}20$ (solid curve) and $\lambda_2\tau_N=0$ (dotted curve) with $T=3{\cdot}75\times10^{-7}$ sec. It is noted that the values of ω_1 and ω_2 for which $R(\omega_m)$ becomes unity are practically independent of the quadrupole interaction parameter λ_2. The determination of H_1 and H_2, for which $R=1$, gives at once μ from eqn. (7). The experiment yielded $H_1=(3030\pm60)$ gauss and $H_2=(6210\pm100)$ gauss, respectively, and the magnetic moment μ of the 247 kev ^{111}Cd state becomes

$$\mu_B=-(0{\cdot}783\pm0{\cdot}028)\mu_0.$$

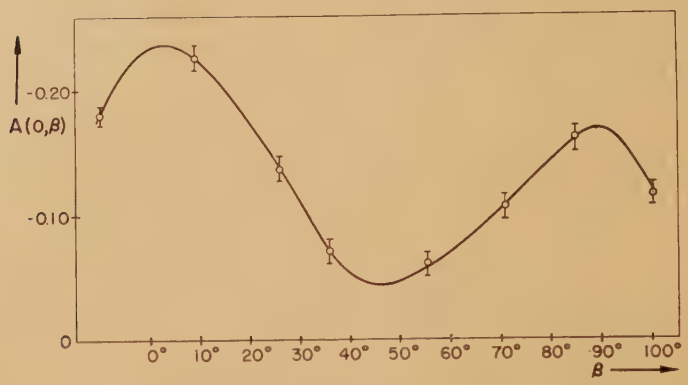
This new value indicates that the magnetic moment of the first excited state of ^{111}Cd is nearer to the Schmitt line than that of the ground state.

It is interesting to note that the experimental points satisfactorily fit the curve $R(\omega_m)$ for $A_2=-0{\cdot}175$ and for a time-dependent quadrupole interaction of $\lambda_2\tau_N=0{\cdot}20$. The latter is of the same order of magnitude as the value extracted from the viscosity dependence of the angular correlation in liquid sources (cf. preceding section).

5.3. *Measurements with* In *Single Crystals as Radioactive Sources*

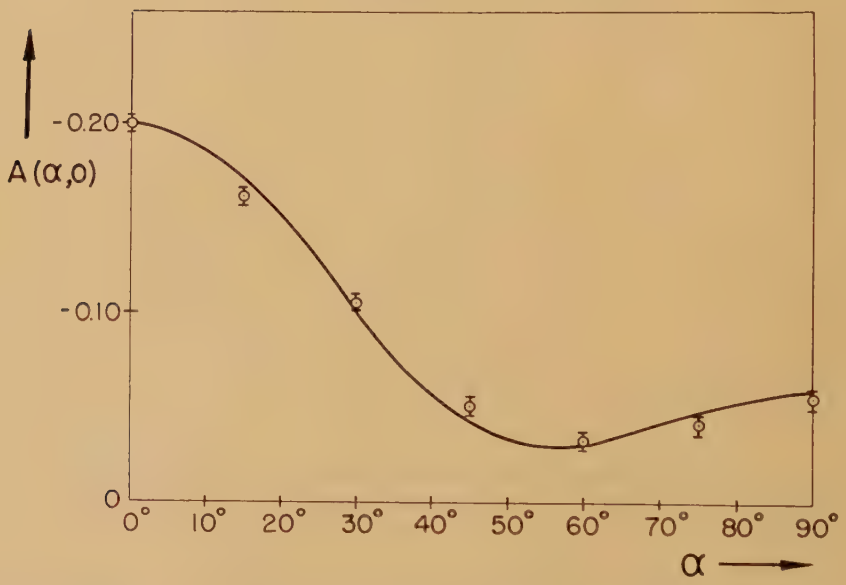
One of the most interesting and conclusive experiments on the attenuation mechanism of the ^{111}In–^{111}Cd gamma correlation are the measurements of the Zürich group performed with In single crystals[55] as sources. The angular correlation displayed by these specially prepared metallic indium single crystals containing radioactive ^{111}In was found to be strongly dependent upon the direction of the crystallographic *c*-axis of the face-centred tetragonal In single crystals, relative to the propagation directions $\mathbf{k}_1$ and $\mathbf{k}_2$ of the two ^{111}Cd gamma rays. The data represented in fig. 21 show the experimental anisotropy values $A(0, \beta)$ for different orientations (angle β) of the *c*-axis in the plane ($\mathbf{k}_1$, $\mathbf{k}_2$) defined by the two detectors and the source (compare fig. 11). The anisotropy as a function of the angle α, the *c*-axis being in the plane defined by the source, the fixed counter and the normal n to the ($\mathbf{k}_1$, $\mathbf{k}_2$) plane, is plotted in fig. 22. Several important conclusions may be drawn from these curves. (i) The single crystal experiments leave little doubt that the attenuating mechanism in metallic In is the static electric quadrupole interaction and

Fig. 21



The anisotropy of the ^{111}In–^{111}Cd gamma–gamma correlation as a function of the angle $\beta(\alpha=0)$ of the crystallographic c-axis of an In single crystal with respect to the propagation direction $\mathbf{k}_1$ (Albers-Schönberg *et al.*, reference (55)).

Fig. 22



The anisotropy of the ^{111}In–^{111}Cd gamma–gamma correlation as a function of the angle $\alpha(\beta=0)$ of the crystallographic c-axis of an In single crystal with respect to the propagation direction $\mathbf{k}_1$. The solid line is the theoretical curve calculated for $\omega_{\rho}{}^{0}\tau_{N}=1{\cdot}60$ (Albers-Schönberg *et al.*, references (55 and 84)).

thus they provide the most convincing proof for the existence of this type of interaction. (ii) The anisotropy measured with the c-axis parallel to one of the propagation directions should correspond to the undisturbed angular correlation if the following conditions are satisfied : no magnetic interaction is present, the In single crystal is without imperfections, and the decay processes leading to the intermediate nuclear state do not alter the environment of the intermediate state of the ^{111}Cd nucleus. These conditions seem to be satisfied and thus the maximum value of the anisotropy $A=-0{\cdot}23$ as measured with these In single crystal sources can, with reasonable certainty, be assumed to be the ' true ' value. The fact that the experimental curves $A(\alpha, 0)$ and $A(0, \beta)$ agree rather well with the theoretical expectations for an axially symmetric electrostatic gradient whose axis is parallel to the crystallographic c-axis indicates that the ^{111}Cd nucleus is retained in a normal lattice position in the indium crystal. Thus the atomic excitation and the recoil in the ^{111}In–^{111}Cd electron-capture decay are apparently not sufficient to break the atom loose from its site in the indium lattice. From a comparison of these experimental results with the theoretical expressions of the angular correlation for pure static quadrupole interaction the Zürich group deduced a value for the interaction parameter[84]

$$\omega_e{}^0\tau_N=\frac{eQ}{2\hbar}\frac{\partial E_z}{\partial z}\frac{3}{(2I_B-1)I_B}\cdot\tau_N=1{\cdot}59\pm0{\cdot}02$$

or for the characteristic quadrupole interaction frequency

$$(eQ/h)(\partial E_z/\partial z)=(6{\cdot}86\pm0{\cdot}4)\text{ Mc/s.}$$

Single crystals of In were also used in an experiment where the angular correlation with magnetic and electric interactions combined was investigated[85] : the anisotropy of a source consisting of an indium single crystal with its c-axis perpendicular to the $(\mathbf{k}_1, \mathbf{k}_2)$ plane was determined as a function of the magnitude of an external magnetic field applied parallel to the c-axis. The preliminary results of this experiment are represented in fig. 23, where the coefficient B_2 of the angular correlation function $W(\Theta)=1+B_2\cos 2\Theta$ is plotted against the magnetic Larmor frequency $\omega_m=\mu H_0/I_B\hbar$. The coefficient B_2 is related to the anisotropy A by $B_2=A/(2+A)$. Again the agreement with theory is remarkably good and the result for the electric quadrupole interaction $\omega_e{}^0\tau_N=1{\cdot}6\pm0{\cdot}2$ deduced from this experiment is consistent with the previously determined more precise value of $1{\cdot}59\pm0{\cdot}02$.

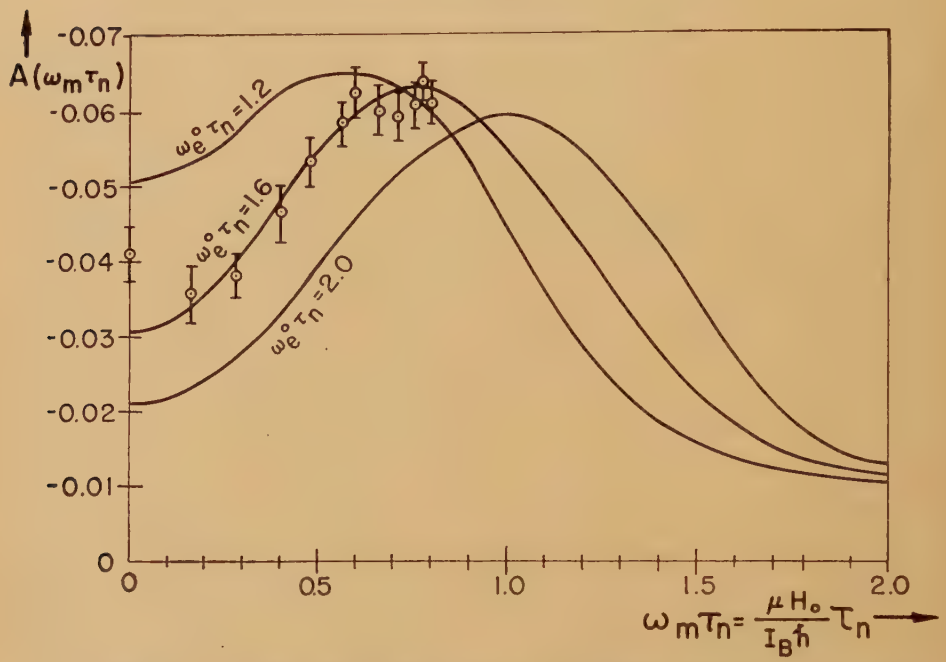
An experimental determination of the attenuation coefficient $G_2(\omega_e{}^0\tau_N)_{\tau_0}$ for the time-integrated angular correlation as measured with sources of polycrystalline metallic In powder provides an additional value for the electric quadrupole interaction in the tetragonal In lattice. Two rather accurate measurements of this coefficient are available[77,86] : $G_2=0{\cdot}37\pm0{\cdot}03$[86] and $G_2=0{\cdot}330\pm0{\cdot}015$[77] corresponding to anisotropy

values of $A=-0{\cdot}095\pm0{\cdot}005$ and $A=-0{\cdot}085\pm0{\cdot}003$ respectively. The characteristic interaction parameter $\omega_e{}^0\tau_N$ is related to G_2 by eqn. (30 *a*) which must be modified to represent the time-integrated attenuation coefficient

$$G_2(\omega_e{}^0\tau_N)_{\tau_0}=\tfrac{1}{35}(7+13I[\omega_e{}^0\tau_N)_{\tau_0}+10I(2\omega_e{}^0\tau_N)_{\tau_0}+5I(3\omega_e{}^0\tau_N)_{\tau_0}]$$

where $I(\omega_e\tau_N)_{\tau_0}$ is defined by eqn. (32). From this expression the values $\omega_e{}^0\tau_N=1{\cdot}4\pm0{\cdot}2$ and $\omega_e{}^0\tau_N=1{\cdot}6\pm0{\cdot}1$, respectively, are deduced.

Fig. 23



The coefficient B_2 of the ^{111}In–^{111}Cd angular correlation function as a function of the magnetic interaction $\omega_m\tau_N$ for the case of the (perpendicular) external magnetic field H_0 being parallel to the symmetry axis of the electrostatic gradient. The open circles represent the preliminary measurements by Albers-Schönberg *et al.*, reference (85)).

An estimate of the gradient of the electric field at the site of the ^{111}Cd nucleus in the tetragonal indium lattice has been made by R. Leidenix[87]: $\partial E_z/\partial z=5{\cdot}8\times10^{16}$ volts cm^{-2}. This calculation, together with the measured quadrupole coupling constant, gives a value for the electric quadrupole moment of the 0·247 Mev excited state of ^{111}Cd $Q\simeq5\times10^{-25}$ cm^2 [84]. Unfortunately the calculation of $\partial E_z/\partial z$ in a metal can be nothing but an estimate and one might think of measuring the value of $\partial E_z/\partial z$ by nuclear induction using a known quadrupole moment as sonde. However, nuclear induction experiments in metals are very difficult to perform

because of the skin effect and furthermore the environment (charge distribution) of the ^{111}Cd impurity in the In lattice might be different from the environment of a normal In nucleus in view of the different electronic structure of the Cd impurity and in view of the preceding In–Cd decay.

5.4. *The Delayed Angular Correlation of the* ^{111}Cd *Gamma Rays*

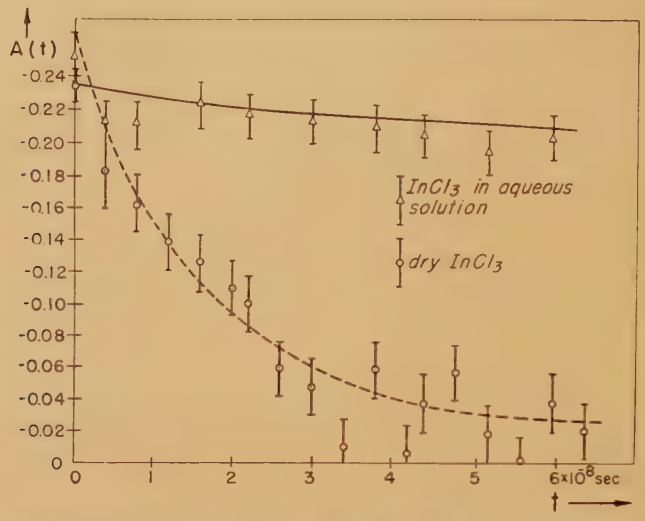
It was pointed out previously (cf. § 4.1) that the nucleus in its intermediate state must be exposed to the interacting fields for a sufficiently long time t in order to observe an appreciable perturbation on the angular correlation. In fact the order of magnitude of the attenuation is $(\omega t)^2$ for $\omega t < 1$. In most practical cases t is essentially determined by the nuclear lifetime τ_N. If, however, the angular correlation is observed with a coincidence analyser, recording the two cascade radiations within a resolving time τ_0 which is much shorter than the nuclear lifetime τ_N ($\tau_0 \ll \tau_N$), then the attenuation of the measured correlation is of the order of $\omega\tau_0 \ll \omega\tau_N$. Provided τ_0 can be made short enough, that $\omega\tau_0 < 0{\cdot}1$, the attenuation of the angular correlation becomes unobservably small, even if the integral correlation, where $\tau_0 \gg \tau_N$, is considerably perturbed.

Such experiments have been recently reported by Fraser and Milton[88]. With a resolving time of $\tau_0 = 1{\cdot}5 \times 10^{-8}$ sec the anisotropy of the ^{111}In–^{111}Cd gamma correlation has been observed. In this fashion polycrystalline $InCl_3$ and In_2O_3 sources exhibited anisotropy values of $A = -0{\cdot}27 \pm 0{\cdot}04$ and $A = -0{\cdot}24 \pm 0{\cdot}04$, respectively. By way of contrast the anisotropies for the integral ($\tau_0 \gg \tau_N$) correlation (table 3) were much smaller for the same sources : $A = -0{\cdot}012 \pm 0{\cdot}005$ and $A = -0{\cdot}085 \pm 0{\cdot}005$ for the $InCl_3$ and In_2O_3, respectively. Also, a dilute aqueous solution of $InCl_3$ yielded a somewhat larger anisotropy ($-0{\cdot}26 \pm 0{\cdot}02$) than did the integral correlation measurement ($-0{\cdot}225 \pm 0{\cdot}001$). The data on the delayed angular correlation (fig. 24) of the aqueous $InCl_3$ solution are compatible with the slow exponential decrease of the attenuation coefficient (eqn. (36)), which is expected as a result of the time-dependent quadrupole interaction in a liquid of small viscosity. The solid curve in fig. 24 was calculated on the basis of the root mean square value of the quadrupole interaction frequency in a liquid as determined by Hemmig and Steffen[79]. The anisotropy of the solid sources, $InCl_3$ and In_2O_3, decreases rapidly with increasing delay time (figs. 24, 25) in qualitative agreement with the expected behaviour of $G_2(t)$ for static or time-dependent interactions. Unfortunately a theoretical treatment of the interaction mechanism(s) in sources of this kind (insulator, non-stationary atomic shell) is lacking at present, and no further information on the interaction parameters can be obtained from these interesting results on the delayed angular correlation.

The rapid decrease of the anisotropy, within 5×10^{-8} sec delay time, and the constancy of the small anisotropy thereafter, explain the failure of the earlier experiments[62] to detect an appreciable change of the

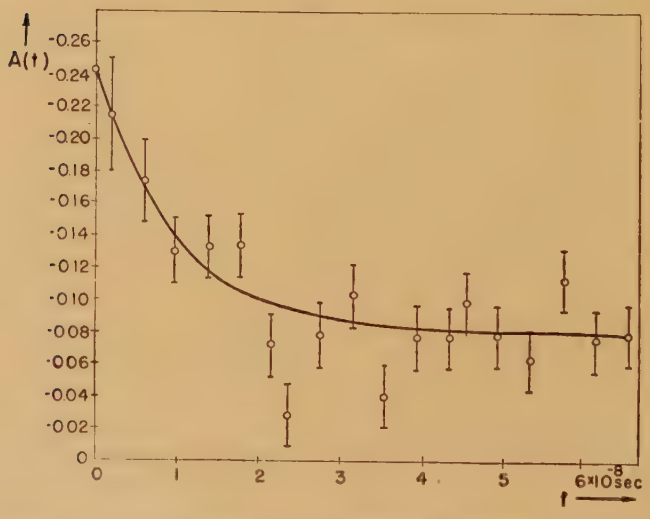
angular correlation by introducing a delay of $1{\cdot}3\times10^{-7}$ sec into one channel of a coincidence analyser which had a resolving time τ_0 as large as $1{\cdot}2\times10^{-7}$ sec.

Fig. 24



The anisotropy of the delayed ^{111}Cd gamma–gamma correlation with sources of solid $InCl_3$ and $InCl_3$ in an aqueous solution as function of the delay time t (Fraser and Milton, reference (88)).

Fig. 25



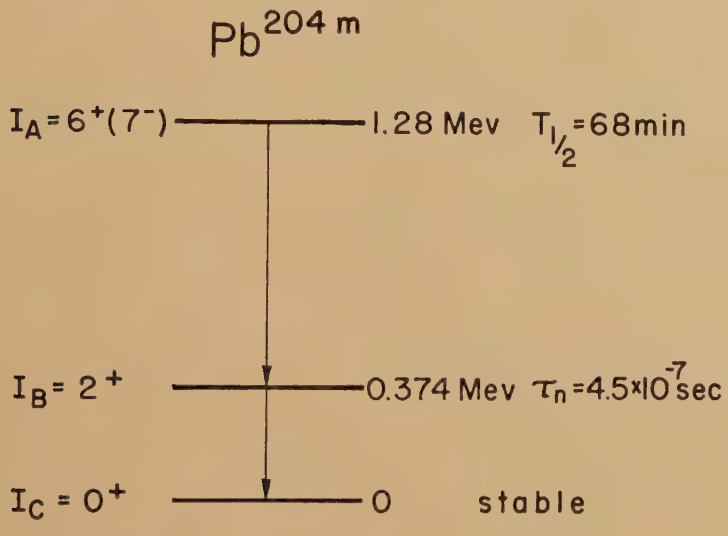
The anisotropy of the delayed ^{111}Cd gamma–gamma correlation with an In_2O_3 source as function of the delay time t (Fraser and Milton, reference (88)).

In summarizing, it can be stated that the experimental results on the ^{111}Cd gamma–gamma cascade allow a consistent, although sometimes only qualitative, interpretation on the basis of the theoretical considerations presented in Part I.

§ 6. The Gamma–Gamma Correlation of the ^{204m}Pb Isomer

The decay of the 68 min isomeric state of ^{204}Pb (fig. 26) has been extensively investigated by the Brookhaven group[63]. The fact that no decay processes which might introduce uncontrollable changes of the nuclear environment precede the gamma–gamma cascade, together with the magnitude of the lifetime of the intermediate state ($\tau_N = 4{\cdot}5 \times 10^{-7}$ sec),

Fig. 26



The decay of the 68 min ^{204}Pb isomer.

makes the ^{204}Pb cascade eminently suitable for investigation of the influence of extranuclear fields. Furthermore, the possibility of measuring the nuclear moments of the first excited state of this nucleus is of particular interest in view of the fact that ^{204}Pb is a nucleus with even numbers of neutrons and protons. In addition, the proton number 82 is a ' magic ' number : the shell of the protons is closed.

The angular correlation of the two ^{204}Pb gamma rays was measured by the Brookhaven group[63] using delayed coincidences ($t = 5 \times 10^{-7}$ sec, $\tau_0 = 2 \times 10^{-7}$ sec) with ^{204}Pb sources in the form of an aqueous lead acetate solution. An anisotropy of $0{\cdot}22 \pm 0{\cdot}05$ was observed in this way. This was the first experimental evidence that some ' memory ' of the angular momentum orientation in the intermediate nuclear state is retained for a time at least as long as 5×10^{-7} sec. The Brookhaven group also

considered the possibility of determining the magnetic moment of the intermediate ^{204}Pb state by measuring the attenuation in an external magnetic field.

Systematic investigations of the influence of the chemical composition and of the physical structure of the radioactive sources on the ^{204}Pb gamma–gamma correlation were made by Frauenfelder, Lawson, Jentschke and de Pasquali[89] at the University of Illinois. Some of their results with liquid and frozen ($T = -185$°c) sources are summarized in table 6. The data obtained with ^{204}Pb sources in the liquid state again show the tendency of liquids to display a much less attenuated correlation than do solid sources. But contrary to the observations with the ^{111}In–^{111}Cd correlation, where all liquid sources exhibited essentially the same, almost unperturbed, angular correlation, the attenuation of the ^{204}Pb correlation is not the same in different liquids, and, in some cases, even apparently identical liquid sources yielded quite different anisotropies, e.g. $Pb(NO_3)_3$ in HNO_3 and $Pb(H_2SO_4)_2$ in H_2SO_4. Frauenfelder *et al.* found consistently high values only with H_2PbCl_6, where the Pb is tetravalent. As pointed out by the investigators this may be due to the fact that the Pb in $(PbCl_6)^{=}$ has coordination number six, and the Pb ion is surrounded by the six Cl atoms in an octahedral configuration. This highly symmetrical environment is expected to result in a relatively small electrostatic gradient at the site of the Pb nucleus.

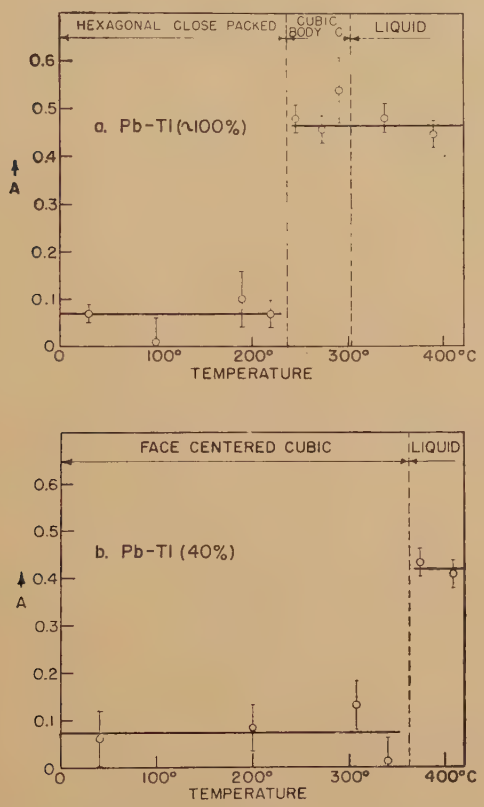
Table 6. Anisotropy Values of the ^{204m}Pb Gamma Cascade with Different Sources (Frauenfelder, Lawson, Jentschke, and De Pasquali. 1953, *Phys. Rev.*, **92,** 1241

Chemical Compound	Solvent	Anisotropy	
		$T = -185$°c	$T = +25$°c
$Pb(NO_2)_2$	HNO_3(1N–16N)	$0{\cdot}08 \pm 0{\cdot}04$	$0{\cdot}30 \pm 0{\cdot}03$
		$0{\cdot}22 \pm 0{\cdot}03$	$0{\cdot}42 \pm 0{\cdot}03$
$Pb(CH_3CO_2)_2$	NH_4 acetate, sat.	$0{\cdot}10 \pm 0{\cdot}05$	$0{\cdot}38 \pm 0{\cdot}03$
$Pb(H_2SO_4)_2$	H_2SO_4		$0{\cdot}23 \pm 0{\cdot}05$
			$0{\cdot}44 \pm 0{\cdot}03$
H_2PbCl_6	HCl12N		$0{\cdot}26 \pm 0{\cdot}04$

To study the effects of crystalline structure the Illinois group measured the ^{204}Pb angular correlation with various Pb–Tl substitutional alloys[90]. The crystalline structure of the mixed Pb–Tl lattices depends markedly upon the relative Pb content of the alloy crystals and on the temperature[91]. Crystals with vanishingly small Pb content have a hexagonal close-packed lattice below the transition temperature of 235°c. Relatively large inhomogeneous electric fields are expected in such a lattice

and indeed a much attenuated angular correlation was displayed by a source of this kind: $A=0{\cdot}07\pm0{\cdot}02$ (fig. 27). Above the transition temperature the alloy changes to a cubic body-centred crystal structure and the anisotropy of the ^{204}Pb correlation rises considerably to the maximum value of 0·47. The same large anisotropy is also found in the liquid phase above the melting point (303°C) of the cubic phase. Quite a different result is found with solid sources of a 60% Pb–40% Tl alloy.

Fig. 27



The influence of the crystalline structure of Pb–Tl alloys on the angular correlation of the ^{204}Pb gamma cascade: (*a*) 100% Tl, (*b*) 60% Pb+40% Tl (Frauenfelder *et al.*, reference (90)).

Up to the melting point (365°C) this alloy has a face-centred cubic crystal structure, for which no electric gradients are expected. Nevertheless, the ^{204}Pb angular correlation displayed by such sources is very strongly attenuated: $A=0{\cdot}07\pm0{\cdot}03$. Only in the liquid phase is the anisotropy of this alloy near the maximum value.

The fact that the Pb–Tl alloys are very probably disordered alloys, in which the Pb sits at random lattice points, might be blamed for the small anisotropy found with the crystalline 60% Pb–40% Tl sources. Although the crystallographic structure of this alloy has cubic symmetry, this symmetry is lacking on a submicroscopic scale if a few of the nearest neighbours of an individual Pb atom are Tl atoms. If the electric charge distributions of the two atoms in the alloy are sufficiently different, there may be present large electric gradients[92], which in general will not be axially symmetric. In contradistinction, in the alloys with vanishingly small Pb content the few Pb atoms are, with great probability, surrounded by Tl atoms only. This more uniform surrounding in a cubic lattice gives, of course, vanishingly small electric gradients and thus could explain the large anisotropy found with these sources.

One serious difficulty in the interpretation of the ^{204}Pb correlation data remains to be explained. The anisotropy values measured with the hexagonal close-packed structure ($0{\cdot}07\pm0{\cdot}01$) and the face-centred cubic disordered alloy ($0{\cdot}07\pm0{\cdot}03$) are considerably below the ' hard core ' values calculated for axially symmetric electrostatic gradients, $A(\infty)=0{\cdot}15$, and for static asymmetric electric gradients, $A(\infty)=0{\cdot}12$, in polycrystalline sources. Thus some time-dependent interaction in addition to the static one seems to be involved in the attenuation mechanism(s) present in these sources. In this connection one might think of an after-effect of the recoil of the ^{204}Pb due to the rather energetic first Pb gamma ray ($E_{\text{recoil}}\simeq 2{\cdot}0$ ev). On the other hand, if this should be a partial cause of the attenuation, the question arises as to why the solid sources of body-centred cubic structure show such a vanishingly small attenuation. A further possibility for the introduction of some time-dependent interaction mechanism is to consider the internal motion of atoms in solids[93–95]. In crystals, which have not absolutely perfect lattices, atoms or molecules occasionally jump from one position to another. The abrupt shift of an atom to a new site in the lattice could give rise to rapidly fluctuating fields at the nucleus. The effect on the angular correlation would be analogous to the motional attenuation mechanism occurring in viscous liquids. However, the frequency ν_j of the elementary jumping process of the diffusion at room temperature seems to be much too small to make it responsible for a time-dependent interaction strong enough to cause the large attenuations observed. In Li metal, e.g., ν_j for self-diffusion is of the order of 10^5 sec^{-1} at room temperature[95]. Activation energies for diffusion in metals are of the order of 10 kcal/mol. Consequently ν_s increases rapidly with temperature, being several thousand times larger at 300°C as compared to room temperature and a strong temperature dependence of the angular correlation should be observed if these ' atomic jumps ' were responsible for the strong attenuation. No such effect has been observed yet and clearly more experimental data are needed before a clarification of the attenuation mechanism involved in solids is possible.

Quite recently the magnetic moment of the intermediate ^{204}Pb state has been determined by applying an external magnetic field to sources which display the unperturbed correlation[83]. The measurements are an exact analogue to the magnetic moment determination discussed for the ^{111}Cd cascade. The result of these measurements,

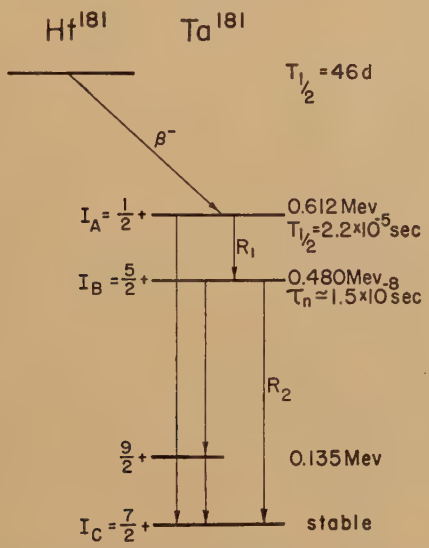
$$\mu_B = +(0{\cdot}14 \pm^{0{\cdot}12}_{0{\cdot}06})\mu_0,$$

although of a preliminary nature, is of particular interest for the discussion of nuclear shell structure.*

§ 7. The Angular Correlation of the ^{181}Ta Gamma Rays

The decay of ^{181}Hf into ^{181}Ta (fig. 28) and the gamma transitions in the ^{181}Ta nucleus, including their angular correlation, have been extensively investigated by McGowan[96]. Angular correlation measurements are somewhat more difficult to perform for the ^{181}Ta gamma rays than for the cases discussed previously, considering the many gamma rays present. The coincidence techniques must be supplemented by gamma-ray spectrometers which allow one to select the gamma rays according to their quantum energy.

Fig. 28



The decay of ^{181}Hf and the gamma transitions in ^{181}Ta (F. K. McGowan, reference (96)). Spin assignments according to reference (97).

Of particular interest is the angular correlation between the 0·132 MeV and 0·480 MeV gamma rays; an intermediate state of relatively long lifetime, $\tau_N = 1{\cdot}5 \times 10^{-8}$ sec, is involved. The angular correlation of this

**Note added in proof.*—Recently Krohn and Raboy [104] discovered that the decay of ^{204m}Pb proceeds by three gamma rays in cascade, instead of two. Nevertheless, our discussion of the extranuclear effects on the angular correlation of the ^{204m}Pb gamma rays is still correct.

cascade is strongly dependent upon the structure of the ^{181}Hf sources used in the measurement, as shown in table 7, which gives the anisotropy values A of the integral correlation ($\tau_0 = 9 \times 10^{-8}$ sec) exhibited by different ^{181}Hf sources. The interpretation of the data should be facilitated by the fact that the gamma cascade of interest follows a metastable state of sufficiently long half-life ($2{\cdot}2 \times 10^{-5}$ sec) to ensure that the environment of the ^{181}Ta nucleus has recovered entirely from the preceding β-decay. Thus no time-dependent interaction mechanism is to be expected in solid ^{181}Hf sources, if the internal motion of atoms in the crystal lattice is neglected (compare § 6). Furthermore, the recoil energy imparted to the ^{181}Ta nucleus by the first gamma ray is small ($E_{\text{recoil}} = 0{\cdot}05$ ev).

Table 7. Influence of Extranuclear Fields on the Angular Correlation of the 0·132 MEV–0·480 MEV Gamma–Gamma Cascade of ^{181}Ta: Anisotropy values observed with ^{181}Hf sources of different chemical composition

Form of the ^{181}Hf source	Anisotropy	Ref.
Hf metal, polycrystalline	−0·08 ±0·01	b
$Hf(OH)_4$, polycrystalline	−0·09 ±0·01*	a
$HfF_4 + HfOF_2$, polycrystalline	−0·085 ±0·010	b
$Hf(OH)_4$, dissolved in 8NHNO$_3$, gelatinous	−0·10 ±0·01*	a
Hf metal extracted into a benzene layer containing thenoyl trifluoro acetone	−0·12 ±0·01*	a
$Hf(NO_3)_4$, dissolved in 8NHNO$_3$, liquid	−0·10 ±0·02*	a
HfF_4 in 0·27NHF	−0·32 ±0·02*	a
HfF_4 in 27NHF	−0·44 ±0·01	a, b
HfF_4 in 27NHF + $HClO_4 . 2H_2O$	−0·38 ±0·02*	a
Same source 24 h after preparation	−0·31 ±0·02*	a
Same source 48 h after preparation	−0·26 ±0·02*	a
Same source 21 days after preparation	−0·22 ±0·02*	a
^{181}Hf in aqu. solution, $t = 27°$C, $\eta \cong 1{\cdot}0$ cp	−0·37 ±0·02	b
^{181}Hf in aqu. solution, $t = 80°$C, $\eta \cong 0{\cdot}5$ cp	−0·42 ±0·02	b
^{181}Hf in aqu. solution + glycerine, $\eta \cong 7$ cp	−0·14 ±0·01	b
^{181}Hf in HCl, $t = 27°$C, $\eta \cong 1{\cdot}5$ cp	−0·23 ±0·02	b
^{181}Hf in HCl, $t = 80°$C, $\eta \cong 1{\cdot}2$ cp	−0·30 ±0·02	b

* Not corrected for the finite angular resolution of the instrument.

a. F. K. McGowan, 1953, *Phys. Rev.*, **93**, 471.

b. H. Paul and R. M. Steffen, *Phys. Rev.*, to be published.

According to McGowan[96] there is evidence that the true unperturbed angular correlation of the two ^{181}Ta gamma rays is given by the function $W(\Theta) = 1 - 0{\cdot}44 P_2(\cos\Theta) - 0{\cdot}10 P_4(\cos\Theta)$ with an anisotropy of $A = -0{\cdot}60$. Hence, even ^{181}Hf sources in form of non-viscous solutions ($\eta \sim 1$ cp) seemed to display an appreciably reduced correlation (table 7). The attenuation coefficients as computed from the correlation measured

with the liquid sources showing the largest anisotropy (Hf in 27NHF) are $G_2=0{\cdot}735$ and $G_4=0{\cdot}497$. The relative values of the two coefficients are consistent with the assumption of a time-dependent quadrupole interaction in liquids (eqn. (38)). Assuming a value of $\tau_c \sim 10^{-11}$ sec for the correlation time in the HfF_4 solutions a value of about 2000 Mc/s is extracted for the root mean square value of the quadrupole interaction $(eQ/h)(\partial E_z/\partial z)$. This value is considerably larger than the corresponding quadrupole interaction frequency in liquid sources of ^{111}In–^{111}Cd. Nevertheless, this would not be surprising, since ^{181}Ta has in its ground state one of the largest electric quadrupole moments known[42], ($Q \simeq 2\times 10^{-24}$ cm^2), and the excited ^{181}Ta states probably have quadrupole moments of the same order of magnitude.

A recent systematic investigation of the time-dependent quadrupole interaction in liquid ^{181}Hf sources of different composition and viscosity, however, seems to indicate a much weaker interaction of the 0·480 Mev ^{181}Ta state with its fluctuating environment, than assumed by McGowan. In fact, if the data measured with sources of widely varying viscosity are extrapolated to zero viscosity, one obtains an unperturbed correlation function $W(\Theta)=1-0{\cdot}29P_2(\cos\Theta)-0{\cdot}07P_4(\cos\Theta)$ with an anisotropy of $-0{\cdot}43\pm 0{\cdot}02$. This value is close to the value which is measured directly with sources of ^{181}Hf in HF, indicating a very small attenuation in this type of source.

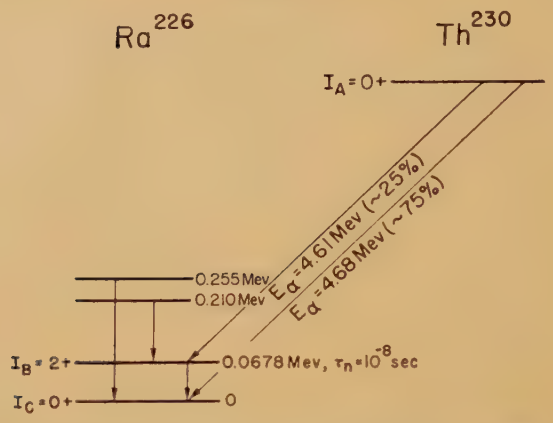
The anisotropy values displayed by polycrystalline Hf metal and HfF_4+HfOF_2 sources ($A=-0{\cdot}08\pm 0{\cdot}01$ and $A=-0{\cdot}085\pm 0{\cdot}010$, resp.) would be below the minimum (' hard core ') anisotropy ($A(\min)=-0{\cdot}125$) allowed for McGowan's unperturbed correlation function. A possible time-dependent interaction mechanism in these solid sources, which could account for the small anisotropy values, is very difficult to visualize in the case of ^{181}Ta. The minimum anisotropy for the extrapolated angular correlation is $A(\min)=-0{\cdot}09$, and is compatible with the observed anisotropy values in solid sources.

The angular momentum assignment based on the extrapolated angular correlation function is quite different from McGowan's and is indicated in fig. 28.

§ 8. The Influence of Extranuclear Fields on Alpha–Gamma Correlations

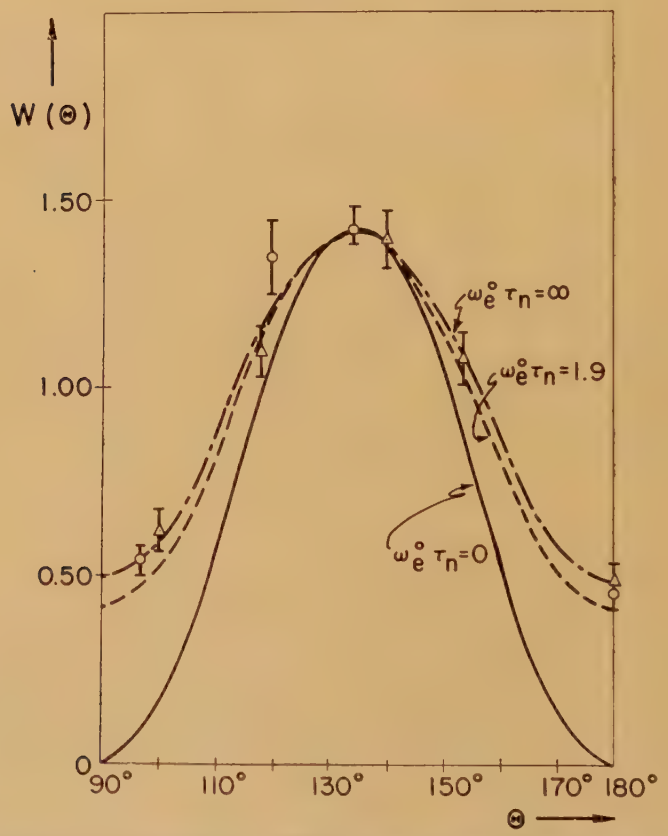
A small percentage of the alpha decay processes leading from ^{228}Th (radiothorium) and ^{230}Th (ionium) to ^{224}Ra and ^{226}Ra, respectively, result in excited states of the daughter nuclei and are followed by emission of low energy gamma radiation (fig. 29). The lifetimes τ_N of the excited states involved in these alpha–gamma cascades are of the order of 10^{-9} sec ; hence sufficiently long that one might expect a perturbation of the alpha–gamma correlation by extranuclear fields. The experimental data for the ^{228}Th alpha–gamma correlation obtained with polycrystalline

Fig. 29



The alpha-decay of ^{230}Th and the gamma transitions in ^{226}Ra. (The alpha-decay of ^{228}Th and the gamma transitions in ^{224}Ra are very similar.)

Fig. 30



The alpha–gamma correlations of ^{230}Th (open circles : Temmer and Wyckoff, reference (101)) and of ^{226}Th (triangles : Battey, Madansky and Rasetti, reference (98)).

sources[98–100] (e.g. $Th(OH)_4$[98]) correspond to an angular correlation function

$$W(\Theta)=1+(0{\cdot}26\pm0{\cdot}02)P_2(\cos\Theta)+(-0{\cdot}87\pm0{\cdot}05)P_4(\cos\Theta)$$

(fig. 30). Similarly, Temmer and Wyckoff[101] found for the alpha–gamma correlation in ^{230}Th (polycrystalline $ThCl_4$ source)

$$W(\Theta)=1+(0{\cdot}27\pm0{\cdot}02)P_2(\cos\Theta)+(-0{\cdot}82\pm0{\cdot}05)P_4(\cos\Theta)$$

(fig. 30). Both correlations show a pronounced departure from the unperturbed alpha–gamma correlation, which for both cases is represented by the function[30,102]

$$W(\Theta)=1+0{\cdot}715P_2(\cos\Theta)-1{\cdot}72P_4(\cos\Theta)$$

(solid line in fig. 30). The values of the attenuation coefficients extracted from these data are $G_2=0{\cdot}37\pm0{\cdot}02$ and $G_4=0{\cdot}49\pm0{\cdot}03$. The fact that G_4 is larger than G_2 excludes the possibility that the attenuation is due to a static magnetic interaction alone, since this type of coupling affects the part of the correlation function represented by $P_4(\cos\Theta)$ more than the one represented by $P_2(\cos\Theta)$ (see fig. 7 of Part I). On the basis of a pure electric quadrupole coupling involving axially symmetric gradients in the polycrystalline sources (eqn. (29), modified for time-integrated correlation) the attenuation is consistent with a value for $\omega_e{}^0\tau_N$ between 1·8 ($G_2=0{\cdot}41$; $G_4=0{\cdot}52$) and $\omega_e{}^0\tau_N=\infty$ ($G_2(\infty)=0{\cdot}37$; $G_4(\infty)=0{\cdot}46$). Unfortunately G_2 and G_4 are rather insensitive to $\omega_e{}^0\tau_N$ for values of $\omega_e{}^0\tau_N>1$ (compare fig. 7). Attempts to use these data to determine or estimate the lower limits of the electric quadrupole moments of the excited ^{224}Ra and ^{226}Ra states fail in view of the impossibility of knowing the electric field gradients at the sites of the Ra nuclei to which a large recoil energy (~100 000 ev) has been imparted by the preceding alpha decay.

Investigations of the alpha–gamma correlation of ^{241}Am with the delayed coincidence technique have been reported recently[88]. When measured with a resolving time considerably shorter than the intermediate lifetime ($\tau_N=10^{-7}$ sec, $\tau_0=1{\cdot}5\times10^{-8}$ sec) the undelayed correlation displayed by polycrystalline Am_2O_3 sources had an anisotropy value of $A=-0{\cdot}15\pm0{\cdot}03$. The insertion of a delay caused a very rapid decrease of the anisotropy and practically no anisotropy was observed at delay times larger than $t=2\times10^{-8}$ sec. Again these results cannot be explained by static interactions alone. Time-dependent interaction mechanisms could be visualized as resulting from the large recoil energy imparted to the residual ^{273}Np nucleus by the alpha particle. The existence of such an attenuation mechanism is not compatible with the observations on the ^{224}Ra and ^{226}Ra alpha–gamma correlation where no evidence of such a time-dependent interaction was found, considering the considerably shorter lifetimes ($\tau_N\simeq10^{-8}$ sec) of the intermediate states involved in the Th–Ra alpha–gamma cascades.

§ 9. Gamma–Gamma Angular Correlations with no Observable Influence of Extranuclear Fields

In as much as the majority of nuclear gamma transitions are dipole or electric quadrupole transitions involving quantum energies of 0·2 MeV or more, it is expected that the lifetimes of the nuclear states from which these gamma rays are emitted are of the order of 10^{-10} sec or shorter. Only in very extraordinary cases could the coupling between nuclear moments and extranuclear fields be so strong as to cause an observable re-orientation of the nuclear angular momentum in such a short time. Thus in most cases $\omega\tau_N < 0{\cdot}1$ for $\tau_N \leqslant 10^{-10}$ sec and no observable influence of extranuclear fields on an angular correlation involving such nuclear states is expected.

Table 8. Anisotropy Values of Various Gamma Cascades observed with Sources in different Chemical and Physical States

Gamma Cascade	Metal	Ionic Crystal	Imbedded in Ag	Dilute Aqueous Solution	Aqu. sol. +glycerine $\eta > 0{\cdot}1$ poise
^{46}Ti	0·165 ± 0·010	0·170 ± 0·005	—	0·165 ± 0·005	0·170 ± 0·005
^{60}Ni	0·167 ± 0·001	0·166 ± 0·003	—	0·167 ± 0·002	0·163 ± 0·003
^{88}Sr	—	−0·095 ± 0·004	—	0·096 ± 0·003	−0·098 ± 0·005
^{106}Pd	0·80 ± 0·02	0·83 ± 0·04	0·81 ± 0·02	0·84 ± 0·02	0·81 ± 0·01
^{114}Cd	0·17 ± 0·01	0·17 ± 0·01	0·16 ± 0·01	0·18 ± 0·01	0·15 ± 0·01
^{122}Te	0·44 ± 0·02	0·46 ± 0·02	—	0·47 ± 0·02	—
^{144}Nd	—	−0·29 ± 0·02	—	−0·29 ± 0·01	−0·30 ± 0·01
^{196}Pt	0·28 ± 0·03	0·26 ± 0·03	—	0·28 ± 0·02	0·29 ± 0·02
^{198}Hg	−0·26 ± 8·02	−0·28 ± 0·02	—	−0·28 ± 0·02	−0·27 ± 0·03

To decide whether or not an angular correlation is attenuated by atomic or crystalline fields it is not sufficient to compare the correlations displayed by different solid polycrystalline sources. In fact, if the correlation is greatly reduced, due to a strong static interaction mechanism present in the solid sources such that $\omega\tau_N \gg 1$, the ' hard core ' correlation is measured which, in general, will be the same for all solid sources. Thus, only if polycrystalline sources as well as liquid sources of different macroscopic viscosity exhibit the same angular correlation is it reasonably safe to assume that the observed correlation is unperturbed by interactions with extranuclear fields.

The results of investigations on some gamma cascades, in which no measurable effects of the chemical composition and structure of the sources could be observed[103], are summarized in table 8.

APPENDIX I

The Representation of the Unperturbed Angular Correlation

The function representing the unperturbed angular correlation can be written in the form of three different finite expansions :

$$W_0(\Theta)=1+\sum_{k=1}^{k_{\max}} A_{2k}P_{2k}(\cos\Theta), \quad . \quad . \quad . \quad . \quad . \quad \text{(A 1)}$$

$$W_0(\Theta)=1+\sum_{k=1}^{k_{\max}} a_{2k}\cos^{2k}\Theta, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(A 2)}$$

$$W_0(\Theta)=\sum_{l=0}^{l_{\max}} b_{2l}\cos 2l\Theta. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(A 3)}$$

The Legendre polynomials $P_{2k}(\cos\Theta)$ are polynomials of degree $2k$ in $\cos\Theta$:

$$\left.\begin{aligned} &P_0(\cos\Theta)=1,\\ &P_2(\cos\Theta)=\tfrac{1}{2}(3\cos^2\Theta-1),\\ &P_4(\cos\Theta)=\tfrac{1}{8}(35\cos^4\Theta-30\cos^2\Theta+3),\\ &P_6(\cos\Theta)=\tfrac{1}{16}(231\cos^6\Theta-315\cos^4\Theta+105\cos^2\Theta-5). \end{aligned}\right\} \quad . \quad \text{(A 4)}$$

They can also be represented as finite Fourier series :

$$\left.\begin{aligned} &P_0(\cos\Theta)=1,\\ &P_2(\cos\Theta)=\tfrac{1}{4}(3\cos 2\Theta+1),\\ &P_4(\cos\Theta)=\tfrac{1}{64}(35\cos 4\Theta+20\cos 2\Theta+9),\\ &P_6(\cos\Theta)=\tfrac{1}{512}(231\cos 6\Theta+126\cos 4\Theta+105\cos 2\Theta+50). \end{aligned}\right\} \quad . \quad \text{(A 5)}$$

In the following the relationships between the coefficients A_{2k}, a_{2k} and b_{2l} are calculated for angular correlation functions involving multipole radiation of order not higher than octupole ($k_{\max}=3$, $l_{\max}=3$).

A_{2k}—a_{2k} :

$$\left.\begin{aligned} A_2&=\frac{0{\cdot}6667a_2+0{\cdot}5714a_4+0{\cdot}4762a_6}{1+0{\cdot}3333a_2+0{\cdot}2000a_4+0{\cdot}1429a_6},\\ A_4&=\frac{0{\cdot}2286a_4+0{\cdot}3117a_6}{1+0{\cdot}3333a_2+0{\cdot}2000a_4+0{\cdot}1429a_6},\\ A_6&=\frac{0{\cdot}0693a_6}{1+0{\cdot}3333a_2+0{\cdot}2000a_4+0{\cdot}1429a_6}. \end{aligned}\right\} \quad . \quad . \quad . \quad \text{(A 6)}$$

$$\left.\begin{aligned} a_2&=\frac{1{\cdot}500A_2-3{\cdot}750A_4+6{\cdot}562A_6}{1-0{\cdot}500A_2+0{\cdot}375A_4-0{\cdot}3125A_6},\\ a_4&=\frac{4{\cdot}375A_4-19{\cdot}688A_6}{1-0{\cdot}500A_2+0{\cdot}375A_4-0{\cdot}3125A_6},\\ a_6&=\frac{14{\cdot}437A_6}{1-0{\cdot}500A_2+0{\cdot}375A_4-0{\cdot}3125A_6}. \end{aligned}\right\} \quad . \quad . \quad . \quad \text{(A 7)}$$

$a_{2k}-b_{2l}$:

$$\left.\begin{aligned} a_2&=\frac{2b_2-8b_4+18b_6}{b_0-b_2+b_4-b_6},\\ a_4&=\frac{8b_4-48b_6}{b_0-b_2+b_4-b_6},\\ a_6&=\frac{32b_6}{b_0-b_2+b_4-b_6}. \end{aligned}\right\} \quad . \ . \ . \quad \text{(A 8)}$$

$$\left.\begin{aligned} b_0&=1+0{\cdot}500a_2+0{\cdot}375a_4+0{\cdot}3125a_6,\\ b_2&=0{\cdot}500a_2+0{\cdot}500a_4+0{\cdot}469a_6,\\ b_4&=0{\cdot}1250a_4+0{\cdot}1875a_6,\\ b_6&=0{\cdot}03125a_6. \end{aligned}\right\} \quad . \ . \ . \quad \text{(A 9)}$$

$A_{2k}-b_{2l}$:

$$\left.\begin{aligned} A_2&=\frac{1{\cdot}3333b_2-0{\cdot}7609b_4-0{\cdot}1909b_6}{b_0-0{\cdot}3333b_2-0{\cdot}0666b_4-0{\cdot}0280b_6},\\ A_4&=\frac{1{\cdot}8266b_4-0{\cdot}9963b_6}{b_0-0{\cdot}3333b_2-0{\cdot}0666b_4-0{\cdot}0280b_6},\\ A_6&=\frac{2{\cdot}2164b_6}{b_0-0{\cdot}3333b_2-0{\cdot}0666b_4-0{\cdot}0280b_6}. \end{aligned}\right\} \quad . \ . \quad \text{(A 10)}$$

$$\left.\begin{aligned} b_0&=1+0{\cdot}2500A_2+0{\cdot}1406A_4+0{\cdot}0977A_6,\\ b_2&=0{\cdot}7500A_2+0{\cdot}3125A_4+0{\cdot}2051A_6,\\ b_4&=0{\cdot}5469A_4+0{\cdot}2461A_6,\\ b_6&=0{\cdot}4512A_6. \end{aligned}\right\} \quad . \ . \quad \text{(A 11)}$$

The anisotropy $A=[W(180°)/W(90°)]-1$, as expressed by the coefficients of the different expansions, becomes :

$$A=\frac{1+A_2+A_4+A_6}{1-0{\cdot}5A_2+0{\cdot}375A_4-0{\cdot}3125A_6}-1, \quad . \ . \ . \quad \text{(A 12)}$$

$$A=a_2+a_4+a_6, \quad . \ . \ . \ . \ . \ . \ . \ . \ . \ . \quad \text{(A 13)}$$

$$A=\frac{b_0+b_2+b_4+b_6}{b_0-b_2+b_4-b_6}-1. \quad . \ . \ . \ . \ . \ . \ . \ . \quad \text{(A 14)}$$

APPENDIX II

The Representation of the Perturbed Angular Correlation

Equation (15) represents the general expression for an angular correlation perturbed by extranuclear fields[46, 56] :

$$W(\theta_1\phi_1\theta_2\phi_2)=\sum_{k_1k_2l_1l_2} I_{k_1}II_{k_2}III^{l_1l_2}_{k_1k_2}Y^{l_1}_{k_1}(\theta_1\phi_1)Y^{l_2}_{k_2}{}^*(\theta_2\phi_2), \quad . \quad \text{(A 15)}$$

where all the parameters responsible for the perturbation of the correlation are contained in the factors $III^{l_1 l_2}_{k_1 k_2}$. The indices k_1 and k_2 are even integers which obey the inequality

$$0 \leqslant k_1 \leqslant 2I_B, \qquad 0 \leqslant k_2 \leqslant 2I_B,$$
$$0 \leqslant k_1 \leqslant 2L_1, \qquad 0 \leqslant k_2 \leqslant 2L_2,$$

where L_1 and L_2 are the highest orders in the multipole expansion of the first and second nuclear radiation, respectively. The coefficients I_{k_1} and II_{k_2} which refer to the first and second nuclear transitions can be calculated from the tables of Biedenharn and Rose[40]. For gamma–gamma correlations involving pure multipole transitions only

$$\left.\begin{aligned} I_{k_1} &= \sqrt{(2k_1+1)} \, . \, F_{k_1}(L_1 I_A I_B), \\ II_{k_2} &= \sqrt{(2k_2+1)} \, . \, F_{k_2}(L_2 I_C I_B). \end{aligned}\right\} \quad . \quad . \quad . \quad . \quad \text{(A 16)}$$

If one of the two gamma transitions is a mixture of L_1 and L_1' radiation with the intensity ratio δ^2 (in Ling and Falkoff's notation[38], $(I_{k_1} II_{k_2})$ can be expressed as

$$\begin{aligned} (I_{k_1} II_{k_2}) = {} & \sqrt{(2k_1+1)}\sqrt{(2k_2+1)} \, . \, F_{k_2}(L_2 I_C I_B) \\ & \times [F_{k_1}(L_1 I_A I_B) + \delta^2 F_{k_1}(L_1' I_A I_B) + (-1)^{I_B - I_A - 1} \\ & \times 2\delta \sqrt{[(2I_B+1)(2L_1+1)(2L_1'+1)]} \, . \, G_{k_1}(L_1 L_1' I_A I_B)]. \end{aligned} \quad \text{(A 17)}$$

The factors F and G are tabulated in Biedenharn and Rose's work. The ordinary spherical harmonics $Y_k^l(\theta, \phi)$ are defined as

$$Y_k^l(\theta, \phi) = P_k^l (\cos\theta) \exp(il\phi),$$

where the $P_k^l(\cos\theta)$ are the associated Legendre polynomials of the first kind

$$P_k^l(\cos\theta) = P_k^{-l}(\cos\theta) = \sin^l\theta \, \frac{d^l}{d(\cos\theta)^l} P_k(\cos\theta).$$

The ordinary Legendre polynomials $P_k(\cos\theta)$ are generally defined as

$$P_k(\cos\theta) = \frac{1}{2^k k!} \frac{d^k}{d(\cos\theta)^k} \sin^{2k}\theta.$$

They are polynomials of degree k in $\cos\theta$ and are normalized such that $P_k(1) = 1$.

Expression (A 15) is valid for the general case of perturbing fields without symmetry axis, e.g. asymmetric electrostatic gradients ($\epsilon \neq 0$), combined magnetic and electric interaction with different directions of the magnetic field and the electric gradient, etc. For interacting fields of higher symmetry expression (A 15) can be simplified considerably.

If the perturbing interaction is such that there exists an axis with the property that the projection of the nuclear angular momentum I_B on

this axis is a good quantum number, then $l_1=l_2=l$ and (A 15) admits of some simplification :

$$W(\theta_1\phi_1\theta_2\phi_2)=\sum_{k_1k_2l} I_{k_1}II_{k_2}\frac{1}{\sqrt{(2k_1+1)}\sqrt{(2k_2+1)}} \times g^l_{k_1k_2}Y^l_{k_1}(\theta_1\phi_1)Y^{-l}_{k_2}(\theta_2\phi_2). \quad . \quad . \quad . \quad \text{(A 18)}$$

This expression is valid for static interactions with fields of axial symmetry, e.g. pure magnetic interaction, pure electric quadrupole interaction with axially symmetric gradients, combined magnetic and electric interactions with both field axes parallel, etc.

The attenuation factors $g^l_{k_1k_2}$ are, in general, complex quantities. For equally sensitive detectors or for pure quadrupole coupling, however, the $g^l_{k_1k_2}$ become real : $g^l_{k_1k_2}\to G^l_{k_1k_2}$. For the delayed angular correlation they can be written as

$$G^l_{k_1k_2}(t)=\tfrac{1}{2}\sum_m S^{k_1k_2}_{ml}[\cos(m\omega_e{}^0t+l\omega_m t)+\cos(m\omega_e{}^0t-l\omega_m t)]. \quad . \quad \text{(A 19)}$$

The coefficients $S^{k_1k_2}_{ml}$ are tabulated for all interesting cases in the theoretical work by Alder *et al.*[56]. The attenuation factors $G^l_{k_1k_2}$ for the time-integrated correlation are easily obtained from eqn. (A 19) by carrying out the integrations (compare eqns. (11) and (12)) or by replacing the cos Ω terms by $I(\Omega)_{\tau_0}$ (eqn. (32)) and $I(\Omega)_\infty$ (eqn. (33)), respectively.

If the energy splitting caused by the perturbing interaction is equidistant, e.g. external magnetic field,

$$g^l_{k_1k_2}=\delta_{k_1k_2}g^l_{k_1k_2}=g^l_{2k} \quad (2k=k_1=k_2, \text{ where } k \text{ is an integer}) \quad . \quad \text{(A 20)}$$

and a further simplification of the angular correlation function becomes possible :

$$W(\theta_1\phi_1\theta_2\phi_2)=\sum_{kl} I_{2k}II_{2k}g^l_{2k}Y^l_{2k}(\theta_1\phi_1)Y_{2k}{}^l(\theta_2\phi_2). \quad . \quad . \quad \text{(A 21)}$$

For an external magnetic field the coefficients g^l_{2k} are quite simple :

$$g^l_{2k}(t)=\exp(il\omega_m t). \quad . \quad . \quad . \quad . \quad . \quad \text{(A 22)}$$

If the axis of *any* interacting field with axial symmetry is perpendicular to the plane (k_1k_2), that is, if $\theta_1=\pi/2$ and $\theta_2=\pi/2$ (compare fig. 6), one has

$$Y^l_{k_1}(\tfrac{1}{2}\pi,\phi)=P^l_{k_1}(0)\exp(il\phi)=C^l_{k_1}\exp(il\phi), \quad . \quad . \quad \text{(A 23)}$$

where $C^l_{k_1}$ is a constant. The angular correlation function then is a function of the angle $\Theta=\phi_1-\phi_2$ only :

$$W(\Theta)=\sum_{k_1k_2l} I_{k_1}II_{k_2}g^l_{k_1k_2}C^l_{k_1}C^l_{k_2}\exp[il(\phi_1-\phi_2)] \quad . \quad . \quad \text{(A 24 } a\text{)}$$

or

$$W(\Theta)=\sum_l B_l\exp(il\Theta), \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(A 24 } b\text{)}$$

where

$$B_l=\sum_{k_1k_2} I_{k_1}II_{k_2}g^l_{k_1k_2}C^l_{k_1}C^l_{k_2}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(A 25)}$$

In general, no simple relationship between the B_l and the corresponding coefficients b_l of the unperturbed correlation exists, due to the occurrence of interference terms ($k_1 \neq k_2$). These terms disappear in the case of the pure magnetic interaction

$$B_l(t)=\exp(il\omega_m t)b_l. \quad \ldots\ldots \text{(A 26)}$$

In the case of the quadrupole interaction or for equally sensitive counters B_l is real.

If the angular correlation is perturbed by static crystalline fields in a source which consists of an ensemble of randomly oriented microcrystals with no preferred direction in the source considered as a whole, the correlation, eqn. (A 15), must be averaged over all orientations of the interacting fields. By virtue of the orthogonality properties of the spherical harmonics all terms disappear in this process, except those for which $l_1=l_2=l$ and $k_1=k_2=2k$. The correlation function is then of the form

$$W(\Theta)=1+\sum_{k=1}^{k_{\max}} G_{2k}A_{2k}P_{2k}(\cos\Theta) \quad \ldots\ldots \text{(A 27)}$$

with

$$G_{2k}=\frac{1}{4k+1}\left[1+2\sum_{l=1}^{2k} G_{2k}^{l}\right]. \quad \ldots\ldots \text{(A 28)}$$

If no interaction is present

$$G_{k_1k_2}^{l}=\delta_{k_1k_2}\,;\; G_{2k}^{l}=1$$

and

$$G_{2k}=1.$$

Thus the unperturbed correlation is given by

$$W_0(\Theta)=1+\sum_{k=1}^{k_{\max}} A_{2k}P_{2k}(\cos\Theta)$$

with

$$A_{2k}=\frac{I_{2k}II_{2k}}{4k+1}. \quad \ldots\ldots \text{(A 29)}$$

The angular correlation function for isotropic sources with time-dependent interactions (e.g. liquid sources) is of the same form as eqn. (A 27) with G_{2k} given by eqn. (36).

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List of Important Symbols

$\mathbf{k}_1$, $\mathbf{k}_2$	propagation directions of nuclear radiations.
Θ	angle between $\mathbf{k}_1$ and $\mathbf{k}_2$.
$W_0(\Theta)$	angular correlation function for an unperturbed correlation.
$W(\Theta)$	angular correlation function for an isotropic source.
$W(\Theta, t)$	angular correlation function for a delayed correlation (isotropic source).
$W_{\tau_0}(\Theta)$	angular correlation function for a time-integrated correlation (isotropic source, coincidence resolving time τ_0).
$W_\infty(\Theta)$	integral angular correlation function (isotropic source, $\tau_0 \gg \tau_N$).
$P_{2k}(\cos\Theta)$	Legendre ploynomial of degree $2k$.
$W(\theta_1\phi_1\theta_2\phi_2)$	general form of an angular correlation function for a non-isotropic source, $\theta_1\phi_1$ ($\theta_2\phi_2$) are angles characterizing the direction of $\mathbf{k}_1$ ($\mathbf{k}_2$), relative to some preferred direction.
$Y_k^l(\theta, \phi)$	ordinary spherical harmonics.
A_{2k}	coefficient (real) of Legendre polynomial in the function for the unperturbed correlation.
a_{2k}	coefficient (real) of $\cos^{2k}\Theta$ in the function for a perturbed or unperturbed correlation.
b_{2l}	coefficient (real) of $\cos 2l\Theta$ or $\exp(i2l\Theta)$ in the function for an unperturbed correlation.
B_{2l}	coefficient (in general complex) of $\exp(i2l\theta)$ in the function for a perturbed correlation (interacting fields perpendicular to plane ($\mathbf{k}_1$, $\mathbf{k}_2$)).
$A(x)$	anisotropy value of an angular correlation, depending upon some interaction parameter x.
g^{2l}	attenuation coefficient (complex) for an angular correlation perturbed by fields perpendicular to the plane (k_1k_2) (multiplies the b_{2l}).
G^{2l}	attenuation coefficient (real) for an angular correlation perturbed by fields perpendicular to $\mathbf{k}_1$ and $\mathbf{k}_2$ for quadrupole interaction or equally sensitive detectors (multiplies the b_{2l}).
$G_{2k}(l)$	attenuation coefficient (real) for the delayed correlation (multiplies the A_{2k}, isotropic source).
$G_{2k}(l)_{\tau_0}$	attenuation coefficient (real) for the time-integrated delayed correlation measured with resolving time τ_0 (isotropic source).
$G_{2k}(\omega\tau_N)_{\tau_0}$	attenuation coefficient (real) for the time-integrated correlation depending on the interaction parameter $\omega\tau_N$ and the finite resolving time τ_0 (isotropic source).
$G_{2k}(\omega\tau_N)_\infty$	attenuation coefficient for the integral correlation ($\tau_0 \gg \tau_N$) depending on the interaction parameter $\omega\tau_N$ (isotropic source).

$G_{2k}(\infty)$	limiting ('hard core') value for a particular static interaction (isotropic source).
$G_{2k}(\min)$	irreducible minimum of the attenuation coefficient for any axially symmetric static interaction in isotropic sources.
$g^{l}_{k_1k_2}$	attenuation coefficient (in general complex) for an angular correlation perturbed by fields with axial symmetry, anisotropic source.
$g^{l_1l_2}_{k_1k_2}$	attenuation coefficient (in general complex) for an angular correlation perturbed by fields without axial symmetry, anisotropic source.
$\mathbf{I}$	nuclear angular momentum vector (dimensionless).
I	nuclear angular momentum quantum number (dimensionless) maximum value of m_x.
I_B	angular momentum quantum number of the intermediate state involved in an angular correlation.
m_x	magnetic quantum number (dimensionless); projection of $\mathbf{I}$ on axis characterized by index x.
$\mathbf{J}$	angular momentum vector of electron shell.
$\mathbf{F}=\mathbf{I}+\mathbf{J}$	vector of total angular momentum of the system nucleus+electron shell.
$\mathbf{S}$	vector of total electron spin.
$\boldsymbol{\mu}_x$	magnetic moment vector of nuclear or atomic state characterized by index x (dimension of magneton).
$\mu_0=e\hbar/2\,\mathrm{Mc}$	nuclear magneton ($5{\cdot}049\times10^{-24}$ erg/gauss).
Q	electric quadrupole moment of nuclear state.
$\mathbf{H}$	magnetic field, atomic or external.
$\mathbf{H}_0$	externally applied magnetic field.
$\partial E_z/\partial z$	electrostatic field gradient with respect to the symmetry axis (z-axis).
ϵ	asymmetry parameter characterizing the deviation of an electrostatic gradient from axial symmetry.
ω_m	angular frequency of Larmor precession.
ω_e	characteristic frequency for electric quadrupole interaction.
$\omega_e{}^0$	fundamental frequency for electric quadrupole interaction (corresponding to smallest energy splitting).
ω_s	characteristic frequency of spin lattice coupling in solids.
λ_{2k}	interaction (relaxation) constant for time-dependent quadrupole interaction.
λ'_{2k}	interaction (relaxation) constant for time-dependent spin–lattice coupling.
τ_N	lifetime of intermediate nuclear state.
τ_0	resolving time of coincidence analyser.
τ_c	correlation time.
t	delay time.
τ_s	relaxation time of electron spin.
T_R	recovery time of electronic shell.

$T_{1/2}$	half-life of a nuclear state.
e	elementary electric charge.
$\hbar=h/2\pi$	Planck's constant, divided by 2π.
M	mass of proton.
k	Boltzmann factor.
T	absolute temperature.
η	macroscopic viscosity of a liquid.

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